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Foreword

The decommissioning of nuclear power stations and their related facilities, as well as the clean-up of sites contaminated by radionuclides from accidents and nuclear weapons programs, are international issues. The remediation of sites at Chernobyl and Fukushima have required and will continue to require decades of effort and billions of dollars. At the center of all of these issues, however, is the storage, transportation and disposal of radioactive waste generated at the back-end of the nuclear fuel cycle. Inaction is not an option because we should not leave a legacy of used fuel, high-level waste and contaminated sites to future generations. Indeed, a failure to solve the nuclear waste problem limits the potential of nuclear power to play a role as a major energy producing system, one that does not produce any significant quantity of greenhouse gases.

Many countries with large volumes of nuclear waste from civilian power production and waste from military programs have started cleaning up contaminated sites and have made significant progress in the design and construction of repositories for permanent disposal of these high-activity wastes. However, progress in other countries has been slow, notably Japan where local opposition has largely halted their waste programs and in the United States, where the demise of Yucca Mountain as a geologic repository for high-level waste and spent nuclear fuel has left the United States without a clear path to the solution of this vexing technical and political problem. The recommendations of the *Blue Ribbon Commission on America's Nuclear Future* have essentially taken the U.S. program back to the first steps of the site selection process.

This volume, edited by three eminent international authorities, is a timely contribution that emphasizes the global nature of the problem. It features contributed chapters by experts from most countries with nuclear programs.

Most importantly, this book highlights the opportunities for good science and engineering that can be applied to some very difficult and complex problems, opportunities that we must address if we are to solve the nuclear waste problems created at the back-end of the nuclear fuel cycle.

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Preface

This book examines the extensive international experience of the management of radioactive wastes. Part I introduces in nine chapters the background to, and principles of, radioactive waste (RAW) management and contaminated site clean-up including waste types and sources of contamination, along with processes and technologies for decommissioning, decontamination and immobilisation as well as consideration of international safety standards. Part II, the bulk of the book, contains 15 chapters summarising the current status of management and clean-up in countries from across the world including a separate chapter covering the Fukushima incident of March 2011. While we endeavoured to cover the whole planet, we were unable to find authors from all countries who were available in the timescale required and so, for example, India is a regrettable omission. Nonetheless, the book presents a thorough and clear view of the international state of play in this area, which is so crucial for the environment and the future of civil nuclear power which it seems can never be separated from politics and the always appropriate but sometimes ill-informed concerns of the public over safety. Thus, for example, England and Wales are covered in a separate chapter from Scotland, whose government has chosen a different path for the management of its wastes. Part III, comprising just three chapters, covers clean-up of sites contaminated by weapons programmes in the USA and the former USSR. Undoubtedly there are others but either we are unaware of them or no-one was willing to provide information about them. While the nuclear community recognises the need for openness and transparency, particularly in the civil nuclear sector, this does not yet, understandably, always fully extend to the military.

Editing a book of this size is an enormous undertaking but it does give the editors an excellent overview and the opportunity to detect key themes in the field. Those which have emerged for us include the importance of developing new waste forms for some of the difficult wastes which to date have been left in the 'too difficult for now, leave until later' category. Plutonium (Pu), iodine (I) and technecium (Tc) are radionuclides which are difficult to incorporate into stable solids and some wastes are ill-defined

so that creativity as well as scientific soundness and engineering pragmatism are needed when developing waste forms to immobilise them. Chapter 6 highlights key new waste form developments using room temperature (non-thermal) and thermal techniques, in particular for production of glass composite materials (GCMs).

Safety is obviously the most important concern when dealing with radioactivity and the need for international agreements and collaborations is crucial, as described in Chapter 3. Underpinning safety, and our understanding of the future behaviour and stability of waste forms, temporary stores and permanent geological disposal facilities, and the transportation mechanisms of radionuclides in the biosphere, is a swathe of computer modelling and performance assessment codes. Developments in theory and simulation and modelling are having significant impact in all areas of technology, and RAW management, with its enormous scales of size, time and complexity, will undoubtedly benefit from these developments.

The book is intended as an introductory overview for post-graduate students and researchers in this field but will also be useful for undergraduates studying physics, chemistry, materials, geography, geology, and environmental or other engineering disciplines with an interest in the welfare of the planet. It will also be a valuable resource for training programmes in new nuclear countries. Inevitable in an edited book with many international authors there are differences of style and approach. There is some repetition between chapters but we believe this is tolerable in order for each to remain as a stand alone resource. We asked each author to include a map of their region to give a better understanding of the geography and to indicate further sources of information for the interested reader. We acknowledge the enormous efforts made by the authors of each chapter and also the team at Woodhead Publishing for their help and support over the three years it has taken to put this book together.

Professor William (Bill) Lee
Professor Michael Ojovan
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1

Fundamentals of radioactive waste (RAW): science, sources, classification and management strategies

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Abstract: Classification systems for the types of radioactive waste (RAW) are described along with sources of controlled wastes (including from power production, military programmes, medical uses and research reactors) and uncontrolled or accidental releases. Options for managing controlled wastes from pretreatment, treatment, conditioning and storage stages through to transportation to final disposal are considered. Immobilisation (wasteform), temporary storage and permanent disposal options including near surface, deep and very deep geological disposal are covered as well as strategies for uncontrolled releases.

Key words: radioactive waste (RAW) classification, temporary storage, geological disposal, immobilisation, uncontrolled releases.

1.1 Introduction

The big issue facing mankind at present is the need for population control. Our complete failure to address it, however, has meant that we are putting increasing pressure on our planet's resources and negatively impacting on our environment. We are striving to fulfil our increasing need for power using a diverse portfolio of means including through nuclear fission. Nuclear fission has provided mankind with a significant proportion of our power for more than 50 years in a far more benign, low carbon and environmentally beneficial manner, and with a significantly lower loss of life in its generation than other sources such as coal, oil and gas. However, mankind has an innate fear of peaceful uses of nuclear energy because of the potential uses of nuclear weapons of devastating destructive ability such as those deployed at Hiroshima and Nagasaki, Japan in 1945. Moreover, mankind fears nuclear accidents, because if they occur, such as those at Chernobyl, Ukraine in 1986 and Fukushima, Japan in 2011, the time it takes to clean up is measured in decades if not centuries.

Sites of underground and above ground nuclear weapons testing and sites of accidental releases from research, manufacturing or storage facilities

have left a complex legacy of contaminated land. Stockpiles of nuclear materials from weapons, submarine reactors and medical isotopes have all been allowed to gather without a definitive disposal disposition. Many of the first generations of nuclear power plant (NPP) have now reached the end of their lives and are in the process of being decommissioned (which again takes many decades). A lack of foresight by those building and designing these reactors, and a lack of political will (and finances) to address the issue of clean-up and waste disposal has meant that programmes to do so have become massive, complex, expensive and high profile. Large-scale decommissioning programmes require a national scale of activity, led by government and overseen by national and international regulators and oversight bodies. They require a coordinated approach and a need to be open with the public and stakeholders affected by the programmes.

Over the last 20 years or so action has begun to be taken. National bodies have been set up to oversee, coordinate and implement decommissioning of NPP and other contaminated sites, to treat, separate and immobilise waste in stable waste forms and to temporarily store in suitable packages and buildings prior to eventual permanent disposal in a geological disposal facility (GDF), also termed a repository. Nonetheless, progress varies from country to country and, from the public's viewpoint, is slow and expensive. In this chapter we introduce the main types of nuclear waste and how they are classified and the major issues in decommissioning and clean-up, including strategies for the management of controlled wastes such as spent fuel (SF) from the open fuel cycle and high level wastes (HLW) from the partly closed fuel cycle as well as strategies for uncontrolled releases.

1.2 Controlled and uncontrolled wastes

Radioactive waste is material that contains, or is contaminated with radionuclides at concentrations or activities greater than the clearance levels set by the regulators, and for which no use is foreseen. The hazard associated with radioactive wastes depends on the concentration and nature of the radionuclides with those emitting higher energy radiation or being more toxic to life, being the most hazardous.

Radiotoxicity is the harmful effect of chemical substances as a result of their containing radioactive elements. The effect of ionising radiation emitted by the elements leads to changes in the metabolism and structure of living organisms. It is a measure of how harmful a radionuclide is to health. The type and energy of rays, absorption in the organism, residence time in the body, etc., all influence the degree of radiotoxicity of a radionuclide.

Alpha particles (He atoms) are very strongly ionising, so if they come into contact with atoms in a living tissue they can cause mutations, unusual

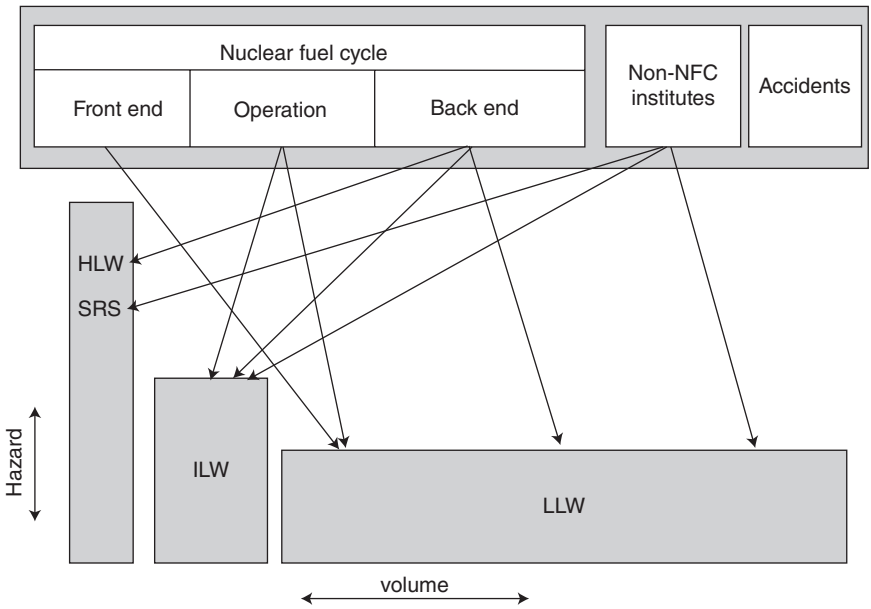
chemical reactions in the cell and possibly cancer. Although the most ionising, it is the least dangerous form of radiation as long as it is not ingested or inhaled, because it is stopped by, for example, a sheet of paper or skin so that it cannot penetrate into your body. Alpha radiation is most commonly used in smoke detectors generated by americium.

Beta radiation is made up of an electron with high energy and speed. Beta radiation is more hazardous because it can also cause ionisation of living cells. Although it is less ionising than alpha radiation, it has the capability to pass through living cells and can be stopped by an aluminium sheet. If beta radiation hits a molecule of DNA it may cause spontaneous mutation and cancer. It is used industrially in thickness measurement such as in paper mills and aluminium foil production.

Gamma rays are high frequency, very short wavelength electromagnetic waves with no mass and no charge. They are emitted by a decaying nucleus so that it can release energy allowing it to become more stabilised as an atom. Gamma rays have the highest penetrating power, only being stopped by a few centimetres of lead or a few metres of concrete. They are the least ionising of the radiations but this does not mean that they are not dangerous. Gamma rays are likely to be emitted alongside alpha and beta radiation but some isotopes only emit gamma radiation. Gamma rays are useful because they can kill living cells and so be used to sterilise by, for example, destroying harmful bacteria. Gamma rays are also used in radiotherapy to kill off cancerous cells. They are also used to sterilise medical equipment, which is particularly useful in tools that would be melted by heat sterilisation or compromised by bleaches and other disinfectants.

Radioactive waste is accompanied by significant levels of radiation, hence it requires not only immobilisation to prevent radionuclides spreading around the biosphere, but also shielding and, in some cases, remote handling. A waste with activity concentrations equal to, or less than, clearance levels is considered non-radioactive. Radioactive wastes are either *controlled* or *uncontrolled*.

Controlled wastes are largely a product of the nuclear fuel cycle (NFC) used to generate electricity for civil use (Fig. 1.1). Wastes are generated during ore mining and processing to access the uranium metal or oxide, its enrichment and synthesis into fuel (the front end of the NFC), the operation and running of the reactor (operations wastes) and from fuel removal, treatment and disposal (the back end of the fuel cycle). Front end waste is contaminated basically with naturally occurring radionuclides, whereas operational waste also contains fission and activated products (typically low level waste (LLW) and to a lesser extent intermediate level waste (ILW); these are defined in Section 1.3). Front end wastes include contaminated mining wastes and uranium hexafluoride tails from enrichment. Operational wastes include spent filters and ion exchange resins, evaporator



1.1 Sources of radioactive waste (adapted from Ojovan and Lee, 2005). HLW = high level waste, ILW = intermediate level waste, LLW = low level waste, SRS = sealed radioactive sources.

concentrates and absorber rods. Back end wastes include sludges from storage ponds, typically cemented ILW and vitrified HLW from reprocessing or spent fuel if direct disposal is planned.

During the early part of the nuclear era, consideration was not given to disposal of radioactive waste. As a result some NFC wastes (now termed legacy or historic wastes) are ill-characterised and stored under conditions which are far from ideal. They comprise a vast range of materials, e.g. Pu-contaminated materials (PCM) such as paper, wood and plastics, fuel cladding, damaged and corroded fuel elements, old tools and equipment and assorted test samples often mixed together. Sometimes these have been stored under water and have degraded over time to form complex sludges and supernatant liquids.

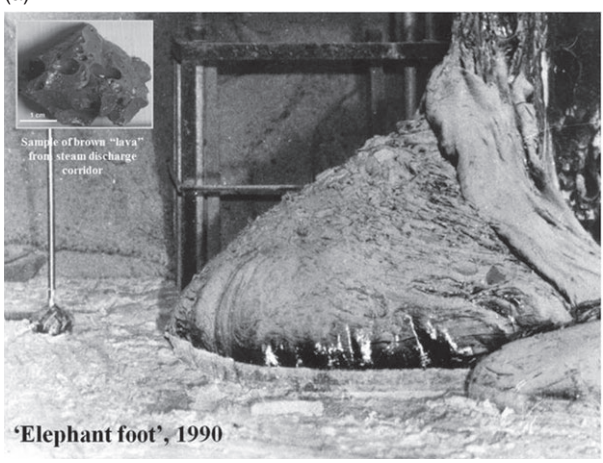
Controlled non-NFC wastes include those from various applications of radionuclides in research, medicine and industry including spent sealed radioactive sources (SRS) of isotopes used in medical applications.

Uncontrolled wastes arise when unexpected events occur or where the level of care was not taken that would be expected today. At Hanford, one of two sites where the US stores its military (defense) wastes, poorly characterised highly active sludges were stored in massive single shell steel tanks

that eventually leaked (Fig. 1.2a). At Sellafield in the UK, some materials were stored in inappropriate open ponds (Fig. 1.2b) where ingress of atmospheric (salty) rain and organic matter (bird droppings, etc.) has added to the complexity of the problem. Uncontrolled wastes also arise from accidents such as at Chernobyl, Ukraine (Fig. 1.2c) and Fukushima, Japan. The financial cost of cleaning up such sites and others where accidental releases of radioactivity have occurred, such as in Fukushima, is enormous. Nonetheless, the nuclear industry is now developing smart clean-up programmes and concepts (Fig. 1.3) and the knowledge gained from these mistakes has helped us be more proactive in dealing with uncontrolled waste.

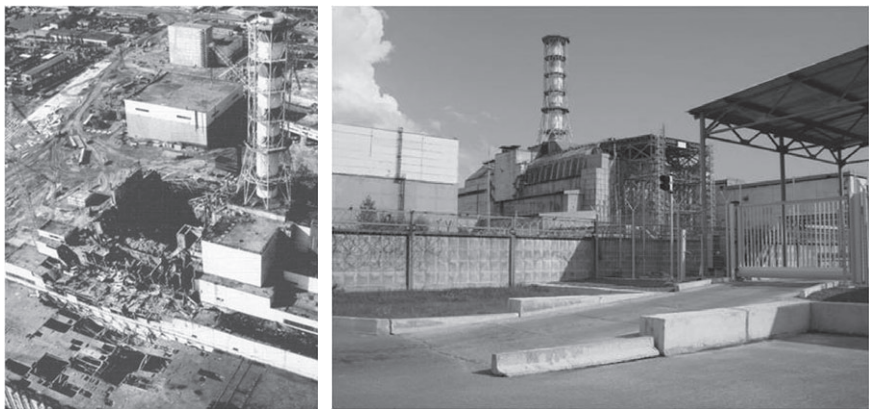
(a)

(b)



(c)

1.2 (a) Early picture of waste containers at Hanford. (b) The open Windscale pile fuel storage pond at Sellafield. (c) Chernobyl NPP lava: a view of highly radioactive so-called 'Elephant foot' and of a sample taken from it (courtesy Boris E. Burakov, Radium Institute, St Petersburg, Russia).



(a) (b)

1.3 The Chernobyl NPP site (a) in 1986 soon after the accident and (b) a current view with protective sarcophagus in place.

Nonetheless, armed with sufficient resources, the results of decades of intensive research and international support progress can be made (Fig. 1.3).

1.3 Radioactive waste (RAW) classification

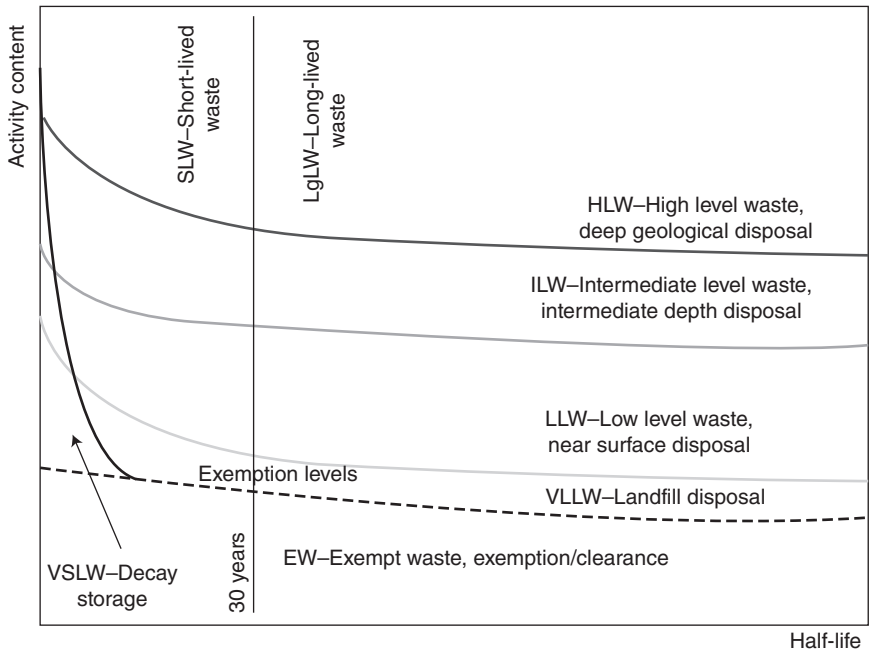
For practical purposes, radioactive waste is classified into different classes depending on actual management needs. A number of parameters are considered in classification schemes, the most important of which are shown in Table 1.1.

Radioactive wastes are typically classified accounting for potential clearance, decay storage or disposal, e.g. final point of waste disposition (IAEA, 2009). Key parameters in the IAEA (International Atomic Energy Agency) classification scheme are radionuclide half-life and radioactivity content. The radionuclides are divided into long-lived and short-lived, where a radionuclide with a half-life longer than that of ^{137}Cs (30.17 years) is considered to be long-lived, whereas those with shorter half-lives are considered short-lived. The activity content is a generic term that covers activity concentration and total activity and is used in classification schemes accounting for the generally heterogeneous nature of radioactive waste (IAEA, 2009). The activity content can range from negligible to very high, e.g. very high concentration of radionuclides or very high specific activity. The radioactivity contents are always analysed compared to exemption levels (IAEA, 2004), e.g. the higher the activity content above those levels the greater the need to contain the waste and to isolate it from the biosphere.

The IAEA classification is shown schematically in Fig. 1.4 and has as lowest by activity content the exempt waste (EW). Exempt waste (EW) is

Table 1.1 Important characteristics of radioactive wastes

Property	Origin	Radiological	Physical	Chemical	Biological
Parameter	Source, Manufacturer	Criticality. Half-life. Heat generation. Intensity of radiation. Activity and concentration of radionuclides. Surface contamination. Dose factors of relevant radionuclides.	Physical state (solid, liquid, gas). Size, volume and weight. Compressibility Dispersibility. Volatility. Solubility. Miscibility.	Potential chemical hazard. Corrosion resistance. Corrosivity. Organic content. Combustibility. Reactivity. Gas generation. Sorption of radionuclides.	Potential hazard. Decomposition rate and products.



1.4 Schematic classification of radioactive wastes aiming for clearance, decay storage or disposal.

that radioactive waste that meets the criteria for clearance, exemption or exclusion from regulatory control for radiation protection purposes which are given in IAEA publications (IAEA, 2003b, 2009). The criteria for exemption were established by the IAEA following the ICRP (International Commission on Radiological Protection) recommendations and principles used to derive exemption levels for radioactive materials. Generically they are based on an expected individual effective dose not higher than 10 μSv/annum and a collective effective dose not higher than 1 person Sv/annum. Exemption levels were established for both concentration and total amount of radionuclides based on the individual and collective dose. These were determined for each radionuclide taking account of all possible pathways to humans including assessment of individual and collective doses. Exemption levels are published in the International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources (IAEA, 2003b). Sources of radiation are exempt from control if at a distance of 0.1 metres, the dose rate is below 1 μSv/h. Clearance levels are defined by the national regulatory authorities; however, since these take into account internationally approved recommendations, quantified clearance levels (with some exceptions) are similar in all countries. EW contains such small concentrations of radionuclides that it does not require

provisions for radiation protection, irrespective of whether the waste is disposed of in conventional landfill sites or recycled, so EW is, in practice, considered as a non-radioactive material.

The IAEA classification scheme defines five classes of radioactive waste: very short-lived waste (VSLW), very low level waste (VLLW), low level waste (LLW), intermediate level waste (ILW) and high level waste (HLW).

VSLW is that radioactive waste which can be stored for decay over a limited period of no longer than a few years with subsequent clearance from regulatory control. Clearance is carried out according to existing national arrangements, after which VSLW can be disposed of, discharged or used. VSLW includes waste containing primarily radionuclides with very short half-lives which are most often used for research and medicine.

VLLW is that radioactive waste which does not necessarily meet the criteria of EW, but that does not need a high level of containment and isolation and because of that is suitable for disposal in near surface landfill type facilities with limited regulatory control. Typical VLLW includes soil and rubble with low levels of activity concentration.

LLW has higher activity contents compared to VLLW, but with limited amounts of long-lived radionuclides in it. Such waste requires robust isolation and containment for periods of up to a few hundred years and is suitable for disposal in engineered near surface facilities. LLW covers a very broad range of waste with long-lived radionuclides only at relatively low levels of activity concentration.

ILW is that radioactive waste that, because of its radionuclide content, particularly of long-lived radionuclides, requires a greater degree of containment and isolation than that provided by near surface disposal. However, ILW needs no provision, or only limited provision, for heat dissipation during its storage and disposal. ILW may contain long-lived radionuclides, in particular, alpha emitting radionuclides that will not decay to a level of activity concentration acceptable for near surface disposal during the time for which institutional controls can be relied upon. Therefore ILW requires disposal at greater depths, of the order of tens of metres to a few hundred metres. A precise boundary between LLW and ILW cannot be universally provided, as limits on the acceptable level of activity concentration will differ between individual radionuclides or groups of radionuclides. Waste acceptance criteria for a particular near surface disposal facility depend on its actual design and operation (e.g., engineered barriers, duration of institutional control, site-specific factors). A limit of 400 Bq/g on average and up to 4,000 Bq/g for individual packages for long-lived alpha emitting radionuclides has been adopted in many countries. For long-lived beta and/or gamma emitting radionuclides, such as ^{14}C, ^{36}Cl, ^{63}Ni, ^{93}Zr, ^{94}Nb, ^{99}Tc and ^{129}I, the allowable average activity concentrations may be considerably higher

(up to tens of kBq/g), although they are specific to the site and disposal facility (IAEA, 2009).

HLW is the radioactive waste with levels of activity concentration high enough to require shielding in handling operations and generate significant quantities of heat by the radioactive decay process typically above several W/m^3. HLW can also be the waste with large amounts of long-lived radionuclides that need to be considered in the design of a disposal facility. Disposal in deep, stable geological formations usually several hundred metres or more below the surface is the generally recognised HLW disposal option.

As can be seen, the IAEA classification scheme is rather generic and has no exact limits in defining radioactive waste classes. Existing national regulations give more exact figures (Ojovan and Lee, 2005). In the UK, radioactive wastes are classified as VLLW, LLW, ILW and HLW (Table 1.2).

1.4 Sources of waste

The main sources of radioactive waste are briefly described in the following sections. More detail is found in the following references (IAEA, 1977, 1988, 1992; Donald *et al.*, 1997; Ojovan and Lee, 2005, 2007; Donald, 2010; Jantzen, 2011) as well as the annual symposium proceedings on the *Scientific Basis for Nuclear Waste Management* (Volumes I–XXXVI) published by the Materials Research Society (e.g., Lee *et al.*, 2008).

Table 1.2 Classification of radioactive waste in the UK

Class	Description
VLLW	Wastes which can be disposed of with ordinary refuse, each 0.1 m^3 of material containing less than 400 kBq of beta/gamma activity or single items containing less than 40 kBq.
LLW	Containing radioactive materials other than those suitable for disposal with ordinary refuse, but not exceeding 4 GBq/te of alpha or 12 GBq/te of beta/gamma activity – that is, wastes which can be accepted for authorised disposal at Drigg, Dounreay or other landfill sites by controlled burial.
ILW	Wastes with radioactivity levels exceeding the upper boundaries for LLW, but which do not need heating to be taken into account in the design of storage or disposal facilities.
HLW	Wastes in which the temperature may rise significantly as a result of their radioactivity, so this factor has to be taken into account in designing storage or disposal facilities.

1.4.1 Nuclear power plant operations and decommissioning

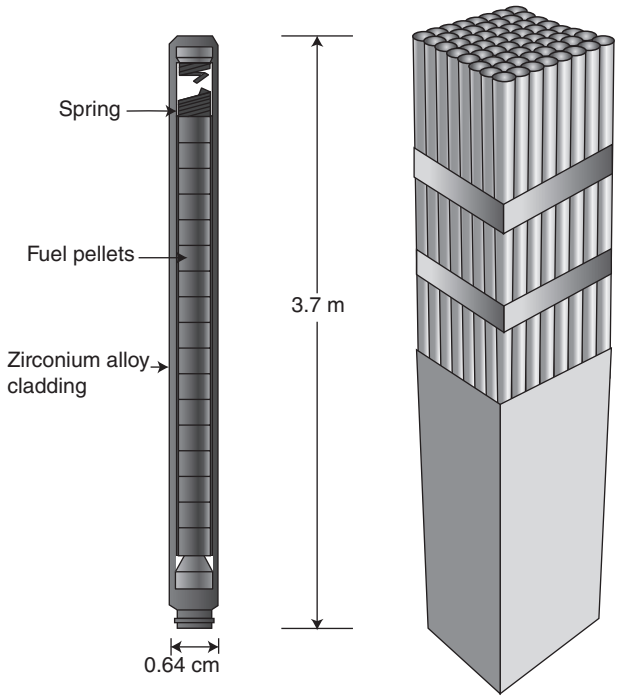
Waste generated during the operation of a NPP is generated mainly by treatment of water from the reactor or ancillaries including SF storage ponds and some decontamination operations. Standard effluent treatment technologies are based on evaporation (distillation), ion exchange, filtration or centrifuging. Typical process wastes from pressurised water reactors (PWR) are borated water concentrates, sludge or filter cartridges, and organic bead resin ion-exchangers (blow-down resins) from primary and secondary circuits, whereas those from boiling water reactors (BWR) are water concentrates and sludge containing different types of ion exchange or filter media as organic powdered resins, diatomaceous earth, activated carbon, cellulose and organic bead resins. Maintenance waste is mainly solid, comprising spent or damaged and contaminated equipment which cannot be repaired or recycled, and items such as contaminated clothes from operators, cardboard, bags, tools and plastic sheeting from maintenance work. Maintenance waste arises also from dismantling the internal structures of the reactor core including the used control rods. Liquid technological wastes comprise mainly oils and small amounts of lubricants and organic solvents used for decontamination. Typically the main radioactive contaminants in operational waste are short-lived radionuclides such as ^{60}Co, ^{90}Sr, ^{134}Cs and ^{137}Cs, although long-lived radionuclides can be present in the internal elements of reactors.

Figure 1.5 shows schematically a nuclear fuel rod and an assembly. The fuel is in the form of ceramic Pu/U oxide pellets in the metal rod. When the fuel reaches the end of its useful life, it is removed from the reactor and is considered as SF. SF contains about 95% ^{238}U, about 3% of fission products and transuranic isotopes, about 1% Pu and 1% ^{235}U.

In the open NFC, the SF is considered as waste and can itself serve as a final waste form since it is a reasonably stable solid providing it is encapsulated in an additional immobilising barrier such as a corrosion-resistant copper or lead container. The ceramic UO_2 matrix of nuclear fuel retains the radionuclides and non-volatile fission products in its open fluorite crystal structure and its polycrystalline microstructure. The metal Zircaloy cladding of the fuel also, if intact, provides an additional barrier. About 30 tonnes of spent nuclear fuel (SNF) waste are typically produced per year by a typical 1 GW NPP.

1.4.2 Reprocessing and recycling facilities

In the closed NFC, SF is reprocessed to extract useful uranium and plutonium. Several reprocessing facilities are currently in operation worldwide,



1.5 Nuclear fuel rod (left) and assembly (right).

including those at Sellafield (UK), La Hague (France) and Chelyabinsk (Russia). These were initially set up to extract material for weapons programmes, but the Pu and U can be recycled for use in PWRs as mixed oxide (MOX) fuel.

Reprocessing of SNF involves removal of the fuel rod metal cladding followed by dissolution of the remainder in the nitric acid, followed by chemical solvent extraction of the uranium and plutonium formed during the fuel burn-up process via the Purex (**P**lutonium and **U**ranium **Ex**traction) process. The remnant solution is HLW and contains the dissolved fission products together with impurities from the cladding materials, inactive process chemicals, transuranic elements formed by neutron capture, and traces of unseparated plutonium. HLW is concentrated by evaporation to reduce the volume and stored in aqueous nitric acid solution in stainless steel tanks. HLWs contain a host of products ranging from uranium fission products to fuel alloying elements, including F, Al, Si and Mo; cladding elements including Zr, Mo, Nb, and Mg; transuranic elements including Np, Am, Cm and residual Pu. HLWs also contain some of the process chemicals including kerosene, tributyl phosphate and related organic materials.

Decommissioning wastes are generated at the end of operation of NFC facilities including nuclear reactors. As well as waste from the radioactive ceramic fuel, some structural materials become activated by elements undergoing neutron capture. The high alloy steel end caps from each fuel bundle in an advanced gas-cooled reactor (AGR), for example, become so activated they are treated as HLW; since there are approximately 100 of these in each AGR assembly and a complete refuel occurs every two years, the amount of waste is significant. The alloying elements of particular concern in steels are Co, Nb, Ni and Mo. After the SNF is removed, the NFC facilities must be decommissioned, demolished and eventually returned to greenfield or brownfield use. During this process, large volumes of waste are generated, although most is not radioactive. The amount of waste arising from decommissioning a typical NPP is 10,000–15,000 tonnes. Much of this waste is concrete and other building material containing only small amounts of radioactivity. About a tenth of the decommissioning waste contains some radioactivity up to the intermediate level.

1.4.3 Research reactors

Many institutions worldwide installed small reactors in support of research and development programmes. Research reactors provide a wide range of training, research, commercial and nuclear power programme support functions from nuclear reactors which are generally not used for power generation. Their output (neutron beams) is used for non-destructive testing, analysis and testing of materials, production of radioisotopes, research and public outreach and education. The UK has had more than 30 research reactors since the 1950s (Table 1.3) but, like all countries, it has shut and decommissioned almost all of them. Its last remaining one Consort, owned and operated by Imperial College London, shut down in December 2012.

Many of these reactors used novel fuels and decommissioning them requires programmes of research to determine suitable ways of managing their wastes. The Nuclear Decommissioning Authority (NDA) in the UK has a special programme examining options for these so-called exotic fuels (NDA, 2012a). For example, such materials held at Dounreay in Scotland include:

- unirradiated plutonium-bearing fuels consisting of plutonium, mixed uranium and plutonium oxide and mixed uranium and plutonium carbide fuels;
- unirradiated high enriched uranium fuels consisting of uranium oxides, uranium metal, uranium alloy, uranium tetrafluoride, uranium hexafluoride and other miscellaneous enriched uranium fuels;

Table 1.3 Current and former research reactors in the UK

Facility name	Thermal power (kW)	Type	Status	Criticality date
Bepo	6,500.00	Graphite, air	Decm	01/01/1962
Berkeley Zero Energy	1.00	Graphite	Decm	14/04/1966
Daphne	0.10	Heavy water	Decm	01/01/1962
Dido	26,000.00	Heavy water	Decm	07/11/1956
Dimple	0.10	Pool	Decm	01/01/1962
Dounreay Fast Reactor	65,000.00	Fast breeder	Shut	01/11/1959
Dounreay MTR	22,500.00	Heavy water	Decm	01/05/1958
Dragon	20,000.00	He cooled	Decm	01/01/1964
Gleep	50.00	Graphite	Shut	18/08/1947
Hazel	0.00	Homog (I)	Decm	01/01/1957
Hector	0.10	Zero power htd	Decm	01/03/1963
Hector	0.10	Graphite CO_2	Decm	10/03/1963
Herald	5,000.00	Pool	Shut	10/10/1960
Hero	3.00	Graphite AGR	Decm	01/06/1962
Horace	0.01	Crit assembly	Decm	01/05/1958
ICI Triga Reactor	250.00	Triga mark I	Decm	01/08/1971
Consort, Imperial College	100.00	Pool	Shut	04/09/1965
Jason	10.00	Argonaut	Decm	30/09/1959
Juno	0.10	Crit assembly	Decm	01/03/1964
Lido	300.00	Pool	Decm	01/09/1956
Merlin	5,000.00	Pool	Decm	01/07/1959
Neptune	0.10	Pool	Oper	03/01/1963
Nestor	30.00	Argonaut	Decm	01/01/1961
Pluto	26,000.00	Heavy water	Shut	25/10/1957
QMC UTR-B	100.00	Argonaut	Decm	10/08/1964
The Univ Research Reactor	300.00	Argonaut	Decm	07/07/1964
UTR-300	300.00	Argonaut	Decm	01/06/1963
Vera Nuclear Assembly	0.10	Crit fast	Decm	01/01/1961
Viper	0.50	Fast burst	Shut	26/05/1967
Vulcan	0.00	PWR	Shut	01/01/1961
Windscale AGR	120,000.00	Graphite AGR	Decm	09/08/1962
Zebra	1.00	Crit fast	Shut	01/12/1962
Zenith I	0.50	Graphite CO_2	Decm	01/12/1959
Zenith II	1.00	Graphite	Decm	01/03/1972
Zephyr	0.00	Crit fast	Decm	01/01/1954
Zeus	0.10	Crit fast	Decm	01/01/1955

Note: Decm = decommissioned.

- irradiated fuels, comprising oxide and carbide fuel consisting mainly of prototype fast reactor (PFR) fuel and the HELIOS material that was irradiated in experimental work.

Credible options for management of these fuels in the near term have been identified but significant R&D may be needed to identify routes to permanent disposal (NDA, 2012a).

Many research reactors use highly enriched uranium (HEU) which, because of its potential to be used in unauthorised nuclear weapons, is a significant security threat. As of 2011, approximately 70 tonnes of HEU is being used in civilian power and research programmes in roughly 30 countries. Securing and eliminating stocks of HEU is necessary to decrease the risk that terrorist groups could use this material to create a nuclear explosion. Some 244 research reactors are in operation or temporarily shut down across 56 countries, while a further 424 reactors have been shut down or decommissioned, and five are planned or under construction.

Many of the research reactors that have been shut down, but not decommissioned, have spent HEU fuel on-site. Threat reduction programmes aimed at reducing the amount of HEU at civilian facilities have been in place since 1978, when the USA initiated the Reduced Enrichment for Research and Test Reactors (RERTR) programme. Russia also initiated a programme to reduce enrichment at Soviet-built research reactors outside the Soviet Union. In the past 25 years, many countries have cooperated with the RERTR programme or initiated their own similar programmes. In 2004, the US Department of Energy (DOE) launched the Global Threat Reduction Initiative (GTRI) which the IAEA, Russia, and others have since joined. Among its goals, the GTRI seeks to 'minimize and eventually eliminate any reliance on HEU in the civilian fuel cycle, including conversion of research and test reactors worldwide from the use of HEU to the use of Low Enriched Uranium (LEU) fuel and targets' (Office of Global Threat Reduction, US National Nuclear Security Administration, http://nnsa.energy.gov).

In addition to converting research reactors that use HEU fuel, the RERTR programme is also working on the conversion of medical isotope producers that use HEU targets in their reactors. The programme includes some of the largest producers of medical isotopes, located in Belgium, the Netherlands and South Africa. Besides converting facilities to use LEU fuel, there have also been efforts to consolidate fresh and spent HEU fuel at a smaller number of relatively secure locations. This has involved removing the fuel, mostly to the US and Russia, from other countries, as well as consolidating the fuel within countries. US programmes in this area have all been subsumed under the 2004 GTRI initiative. Together, the two programmes have returned over 2,000 kg of spent and fresh HEU fuel to the USA and Russia since 2004.

1.4.4 Medical and industrial sources

Types and volumes of waste from applications of radionuclides in research, medicine and industry vary extensively in radiochemical, chemical and physical content. Table 1.4 shows some waste types from non-NFC institutions.

Research establishments are often involved in monitoring the metabolic or environmental pathways associated with materials as diverse as drugs, pesticides, fertilisers and minerals. The radionuclides most commonly employed in studying the toxicology of many chemical compounds and their associated metabolic pathways are ^{14}C and ^{3}H, as they can be incorporated into complex molecules with considerable uniformity. ^{125}I has proved valuable in protein labelling. A spectrum of other radionuclides is available for research. Most of the radioactive waste generated by nuclear research centres contains mainly short-lived radionuclides although long-lived radionuclides such as ^{14}C, fissile radionuclides and transuranic elements may also be present.

The main applications of radionuclides in medicine are in radio-immunoassays, radio-pharmaceuticals, diagnostic procedures and radiotherapy. The radionuclides used in hospitals for medical diagnostic procedures and treatments are very short-lived, and the waste generated is usually stored for decay before further treatment as non-radioactive waste. Positron emission tomography (PET), for example, incorporates cyclotron-generated ^{11}C (20 minute half-life) or ^{18}F (110 minute half-life) in a molecule such as sugar which is intravenously administered to the patient and is detected during its circulation around the body. Some radionuclides used in medical applications, however, have longer half-lives including ^{57}Co (271.7 days) used in clinical measurements and ^{3}H (12.3 years) and ^{14}C (5,730 years) used in radio-labelling (Ojovan and Lee, 2005). Medical applications of radionuclides such as for bone densitometry, manual brachytherapy and whole blood irradiation not only may use small quantities of unsealed sources and liquid solutions, but also of highly radioactive sealed radioactive sources (SRS) housed in shielded assemblies. Spent SRS are extremely hazardous as they may contain large quantities of radionuclides. Programmes to

Table 1.4 Waste types from radionuclide applications

Solid	Aqueous	Organic liquids	Highly active
Metallic scrap, brickwork, sorbents including ion-exchange resins, glassware, filters, cardboard, plastics, paper, swabs, tissues, protective clothes, gloves.	Effluents from laboratories, hot cells, fuel storage pool, sump, decontamination; rinsing waters, mining-milling raffinates.	Pump oils, scintillation liquids, extraction solvents such as tributyl phosphate (TBP), kerosene and amine.	Sealed radioactive sources, radium needles, reactor-irradiated materials.

collect, consolidate, store and dispose of SRS are being developed (Ojovan *et al.*, 2004; IAEA, 2005, 2008a).

1.4.5 Nuclear weapons programmes

Defence-related wastes tend to be simpler than those from commercial nuclear applications. Wastes derived from Pu production contain high levels of sodium, due, for example, to the need to neutralise the acidic liquor before it could be stored in the carbon steel tanks built in the early days of the US defence programme at Hanford and Savannah River. Generally, defence wastes do not contain the high concentrations of fission products found in commercial wastes, the exception being the calcined naval reactor wastes currently stored at the Idaho National Laboratory (INL) but destined for the Waste Isolation Pilot Plant (WIPP) in New Mexico, USA. Donald (2007) gave generic compositions for both commercial and defence wastes (Table 1.5), and although there are very large compositional ranges for the constituents, it does highlight the lower proportion of fission products but higher proportion of actinides present in defence waste.

In addition to the wastes generated from commercial energy supply and during the manufacture of warheads, there is also excess plutonium which has been declared surplus to requirements following the decision by the US and Russia to reduce their warhead stockpiles. Under the 1993 Non-Proliferation and Export Control Policy, the US declared 55 tons of plutonium surplus to national security needs. A similar quantity was also declared

Table 1.5 Generic compositions of typical radioactive wastes (mass%)

Constituent	Commercial waste	Defence waste
Na_2O	0–39	0–16
Fe_2O_3	2–38	24–35
Cr_2O_3	0–2	0–1
NiO	0–4	0–3
Al_2O_3	0–83	5–9
MgO	0–36	0–1
MoO_3	0–35	0–1
ZrO_2	0–38	0–13
SO_4	0–6	0–1
NO_3	5–25	0
Fission product oxides	3–90	2–10
Actinide oxides	<1	2–23
Other constituents	–	17–27

Source: Donald (2007).

surplus by Russia. These quantities may be further increased following the 2010 US–Russia strategic arms reduction agreement. It is planned to utilise this where possible in MOX fuel.

Finally, weapons testing has left a legacy of contaminated sites worldwide. These include Semipalatinsk and West Kazakhstan (Kazakhstan), Novaya Zemlya (Russia), Lop Nor (China), Maralinga (Australia) and others in the Pacific islands, India, Pakistan and Korea. The first atmospheric tests were conducted at the Nevada test site (USA) in 1951. Following the Limited Test Ban Treaty of 1963, atmospheric testing ceased, and nearly 90 percent of the US underground weapons tests were detonated in Nevada. Congress imposed a moratorium on testing of nuclear weapons, and in 1992, underground testing ceased. A total of 907 underground nuclear detonations were conducted above, near and below the groundwater table in alluvial basins, in volcanic highlands, in shafts and tunnels of zeolitised volcanic rocks, and in tunnels mined in granitic rock. Underground testing at Nevada deposited an estimated 132 million curies of radioactivity below ground, decay corrected to 1992. These topics are considered in details in the last three chapters of this book.

An underground explosion produces a spherical cavity from combined vaporisation, melting and shock compression of the rock. As the detonation pressure subsides, the rocks above the cavity typically collapse (timeframe of seconds to days after the test) and the cavity is filled with rubble consisting of collapsed rock, and solidified rock melt (melt glass). The collapse void can propagate upward variable distances forming a chimney that may or may not extend to the surface forming a subsidence crater. The temperature and pressure history of an explosion and response of the surrounding rock control the distribution of radionuclides around the test. Radionuclides produced underground include tritium, fission products, actinides and activation products. Refractory radionuclides are trapped primarily in the melt glass, and in cavity rubble and compressed rock around the cavity; volatile species circulate outward and condense in cracks and void spaces for distances of 1–3 cavity radii from the test point (Pawloski *et al.*, 2008). The extensive contamination of the land at such sites and the potential for spread via local hydrology and hydro-geological has led to extensive studies of such sites (e.g., Busygin *et al.*, 1996; D'Agnese *et al.*, 1997).

1.4.6 Accidental releases

Radioactive contamination and waste may also arise from accidents. Accidents generate radioactive waste of volume and composition which depend on the material involved and the magnitude of the accident. The International Nuclear and Radiological Event Scale (INES) was developed in 1990 by international experts convened by the IAEA and the OECD Nuclear

Energy Agency (OECD/NEA) with the aim of communicating the safety significance of events at nuclear installations (IAEA, 2008b). The INES facilitates understanding, using a numerical rating to explain the significance of nuclear or radiological events in a similar fashion to the Richter scale for earthquakes. INES applies to any event associated with the transport, storage and use of radioactive material and radiation sources. Such events can include industrial and medical uses of radiation sources, operations at nuclear facilities, or the transport of radioactive material. Events are classified at seven levels (Fig. 1.6): Levels 1–3 are 'incidents' and Levels 4–7 'accidents'. These levels consider three areas of impact: people and the environment, radiological barriers and control, and defence in depth. The scale is designed so that the severity of an event is about ten times greater for each increase in level on the scale. Events without safety significance are called 'deviations' and are classified Below Scale/Level 0.

The partial core meltdown accident at Three Mile Island (TMI), Pennsylvania, USA in 1979 was at Level 5 on the INES scale, while those at Chernobyl and Fukushima were Level 7. The proper management of the

1.6 The International Nuclear and Radiological Event Scale (from the IAEA website).

TMI accident meant that there were no person overexposures to radiation and no casualties, so keeping it at Level 5.

Table 1.6 shows the most significant accidents involving radioactive materials. Accidents involving SRS are worryingly common. Over 2,300 cases have been reported of SRS found in scrap metal. A large number of cases have been reported of accidental melting of SRS with scrap metal in, for example, steel and aluminium foundries. The total number of cases of melting SRS with scrap metals exceeds 60 in 18 countries. In Algeciras, Spain in 1998, radioactive gases, aerosols and particles from melting SRS with scrap were released and detected all over Europe. Concentrations up to 2,000 Bq/m^3 of ^{137}Cs in the air were detected although the incident had minor consequences. Since 1983, 30 cases of melting of SRS with scrap metal occurred in the US, which required $8–10 million in each case to decontaminate and restore the metallurgical facilities. In 1987 a serious accident occurred in Goiania, Brazil with a ^{137}Cs SRS left within a teletherapy unit. The SRS was found by two scavengers who took the unit home, removed the source from the unit and ruptured the source capsule. This caused significant contamination of people and the surrounding environment. Four severely exposed people died and the health of many others was seriously affected. More than 112,000 people were monitored for radiation exposure, of which nearly 300 showed ^{137}Cs contamination. The emer-

Table 1.6 Severe accidents involving radioactive materials

Year	Place	Source	Dose	Overexposures/ deaths
1945/46	Los Alamos, USA	Criticality	≤13 Gy	10/2
1961	USSR	Submarine accident	1–50 Gy	>30/8
1961	Idaho Falls, USA	Explosion in reactor	≤3.5 Gy	7/3
1962	Mexico City, Mexico	^{60}Co SRS	9.9–52 Gy	5/4
1963	China	^{60}Co SRS	0.2–80 Gy	6/2
1964	Germany	^{3}H	10 Gy	4/1
1964	Rhode Island, USA	Criticality	0.3–46 Gy	4/1
1984	Morocco	^{192}Ir SRS	Unknown	11/8
1986	Chernobyl, USSR	NPP	1–16 Gy	134/31
1987	Goiania, Brazil	^{137}Cs SRS	≤7 Gy	50/4
1992	China	^{60}Co SRS	>0.25–10 Gy	8/3
1996	Costa Rica	^{60}Co SRS	60% overdose	115/13
2011	Fukushima	NPP	>0.25 Sv	6/none

Source: Adapted from Ojovan and Lee (2005).

gency response and clean-up effort of houses, buildings and land lasted six months.

The Chernobyl accident in 1986 was due to lack of care in operation and disregard for standard safety procedures. The resulting steam explosion and fire released about 5% of the radioactive reactor core into the atmosphere. Some 31 people were killed in the first few weeks after the accident, and there have since been other deaths from thyroid cancer due to the accident. An authoritative UN report in 2000 concluded that there is no scientific evidence of significant radiation-related health effects to most people exposed to radiation during or after the accident.

The most recent accident was that of 11 March 2011 at Fukushima in Japan. A major earthquake, followed by a 15 m tsunami caused the deaths of over 20,000 people and led directly to the shutdown of three reactors and eventually to significant escape of radioactive material to the environment. Three of the Fukushima Daiichi reactor cores were severely damaged in the first three days, releasing high levels of radioactive materials into the land, sea and air environments. The Japanese authorities announced an official 'cold shutdown condition' in mid-December, as reactor temperatures had fallen to below 80°C at the end of October 2011. According to the Japanese government, the total amount of radioactivity released to date is approximately one-tenth that released during the Chernobyl disaster However, the full extent and level of radioactive contamination remain unclear.

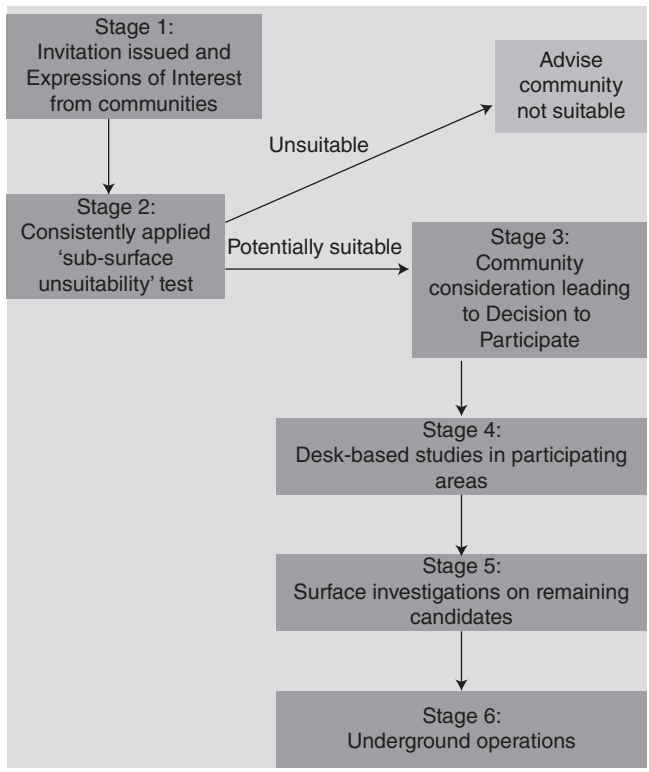
1.5 Managing controlled wastes

1.5.1 Government-led programmes

Mechanisms for managing controlled radioactive wastes are invariably under national government control with legislative and regulatory systems in place to ensure safety and security. In the UK, for example, in 2004 the government commissioned an independent Committee on Radioactive Waste Management (CoRWM) and in 2005 it established the NDA to ensure its 20 civil public sector nuclear sites were decommissioned and cleaned up, safely, securely, cost effectively in ways that would protect the environment for this and future generations. CoRWM recommended to government (CoRWM, 2006) that geological disposal be the end-point for long-term management of RAW but with robust storage in the interim period with provision against delay or failure in reaching the end-point. It also recommended a staged process with flexibility in decision making and partnership with communities willing to participate in the siting process and an expanded national R&D programme to support the process. In response

the government published a White Paper outlining the process and stages (Fig. 1.7) that would lead to permanent geological disposal of the UK's wastes (DEFRA, 2008). Figure 1.7 shows steps in the UK's Managing Radioactive Waste Safely (MRWS) process.

An invitation was sent out to communities in stage 1, inviting expressions of interest in hosting a repository or geological disposal facility (GDF). In stage 2, simple criteria were used to determine if the location was likely to be suitable. At this stage, areas were ruled out, for example, if they had mineral resources which might prove useful in future or aquifers. Communities in potentially suitable areas could decide to participate further in stage 3, while in stage 4 desk-based studies would be carried out which would lead to borehole investigations in stage 5 prior to actual construction of the GDF underground in stage 6. Extensive work is needed during the early stages to underpin the safety case to the regulators to allow construction and safe operation and eventual closure of the GDF, including decades of R&D. This volunteer approach also needs intensive public and stakeholder



1.7 Stages in the UK's Managing Radioactive Waste Safely process.

engagement to convince communities that this is the right approach to dealing with the waste problem. UK government extended the NDA's responsibility to include geological disposal of the waste and in 2007 it established the Radioactive Waste Management Directorate (RWMD) as the implementing body responsible for constructing the GDF.

In other countries the process of developing a strategy for managing radioactive waste has been difficult. In the USA and Japan national programmes have been hindered by a lack of public support, and without a clear end-point (repository site) the programmes flounder. The Yucca Mountain project in Nevada, USA, was hindered by lack of public acceptability, legal challenge and technical shortcomings. In 2009, the Obama Administration announced that it had determined that developing a repository at Yucca Mountain was not a workable option and that the US needs a different solution for nuclear waste disposal. The Secretary of Energy established the Blue Ribbon Commission (BRC) on America's Nuclear Future in January 2010 to evaluate alternative approaches for managing SF and HLW from commercial and defence activities. The BRC conducted a comprehensive review of policies for managing the back end of the nuclear fuel cycle. It has provided recommendations for 'developing a safe long-term solution to managing the Nation's used nuclear fuel and nuclear waste.' Their final report was submitted to the Secretary of Energy in January 2012 (BRC, 2012) and it contained eight recommendations for legislative and administrative action to develop a 'new' strategy to manage nuclear waste:

1. A new, consent-based approach to siting future nuclear waste management facilities.
2. A new organization dedicated solely to implementing the waste management programme and empowered with the authority and resources to succeed.
3. Access to the funds nuclear utility ratepayers are providing for the purpose of nuclear waste management.
4. Prompt efforts to develop one or more geological disposal facilities.
5. Prompt efforts to develop one or more consolidated storage facilities.
6. Prompt efforts to prepare for the eventual large-scale transport of SF and HLW to consolidated storage and disposal facilities when such facilities become available.
7. Support for continued US innovation in nuclear energy technology and for workforce development.
8. Active US leadership in international efforts to address safety, waste management, non-proliferation, and security concerns.

The near-term direction advocated by the BRC aligns with ongoing DOE programming and planning. Current programmes will identify alternatives

and conduct scientific research and technology development to enable long-term storage, transportation, and geological disposal of SF and all radioactive wastes generated by existing and future NFCs. The BRC report has informed the Administration's work with Congress to define a responsible and achievable path forward to manage SF and nuclear waste in the US. The US DOE endorsed the key principles of the BRC recommendations and published a strategy (DOE, 2013) to move forward with their implementation.

1.5.2 Management prior to disposal

While the above highlights the need for a clear end-point (permanent geological disposal), political will and public support, much radioactive waste management must be done prior to disposal. Radioactive waste management approaches vary from country to country. However, a key aspect is to know what waste you have. A national inventory must be collected as is done in the UK (NDA, 2010a) and all other countries.

Figure 1.8 shows a flowchart for solid radioactive waste management prior to disposal, i.e., pre-disposal (Ojovan, 2011). Figure 1.9 reveals that all activities concerned with radioactive waste are conventionally divided into pre-disposal and disposal stages.

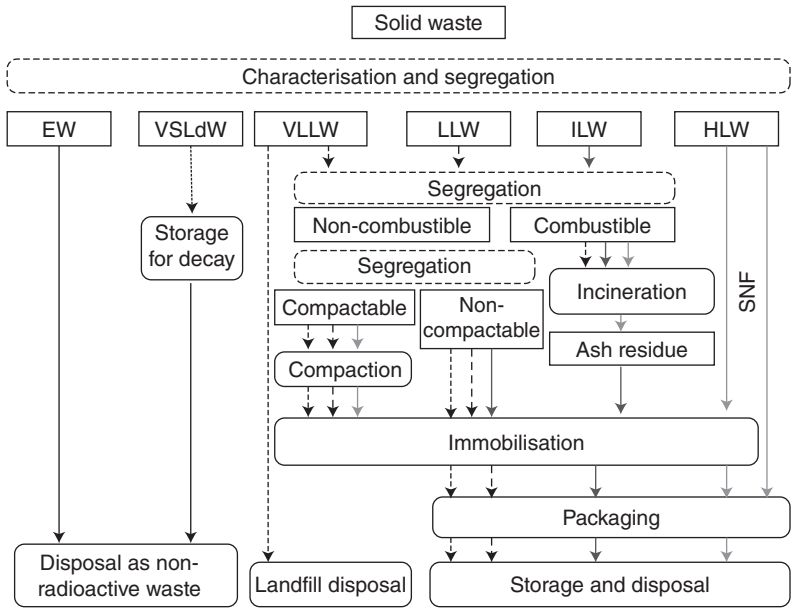
Disposal is the final step in managing radioactive wastes whereas pre-disposal includes activities such as decommissioning, pre-treatment, treatment, conditioning, immobilisation, storage and transport. While various disposal options are available, it is most likely that immobilised wastes will be disposed of in GDFs of one sort or another.

Waste management requires a series of steps:

- pursuing opportunities for waste minimisation
- re-use and recycling
- waste treatment
- packaging
- storage
- transport and then final disposal where required.

This waste hierarchy indicates the preferred options in the managing of waste where disposal is very much the last option; it can be represented as in Fig. 1.10.

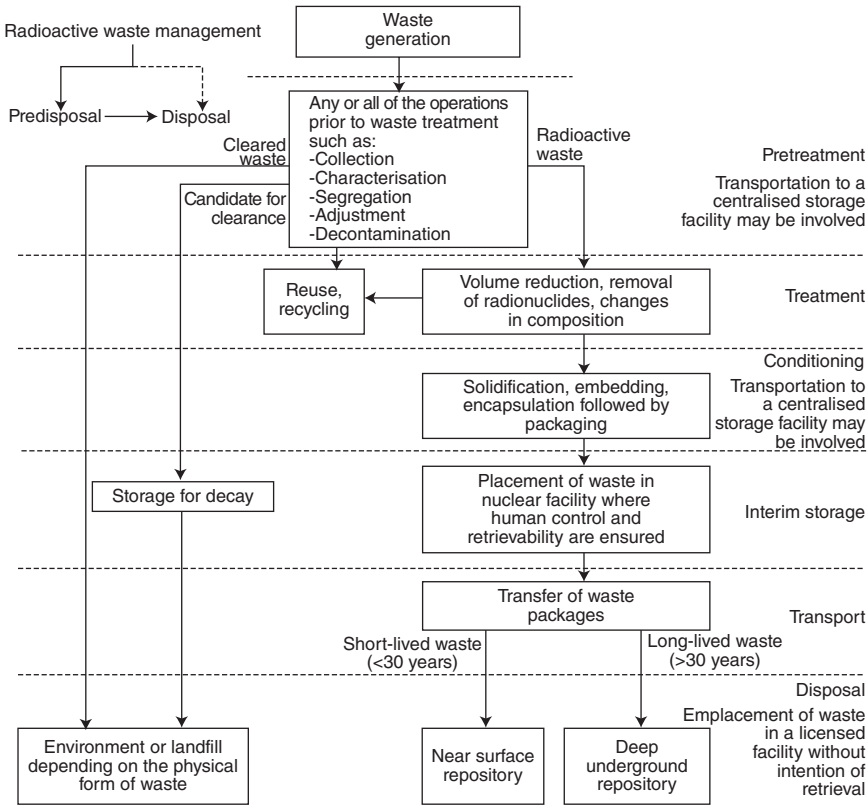
Waste minimisation is a process of reducing the amount and activity of waste materials to a level as low as reasonably achievable. Waste minimisation is now applied at all stages of nuclear processing from power plant design through operation to decommissioning. It consists of reducing waste generation as well as recycling, reuse and treatment, with due consideration



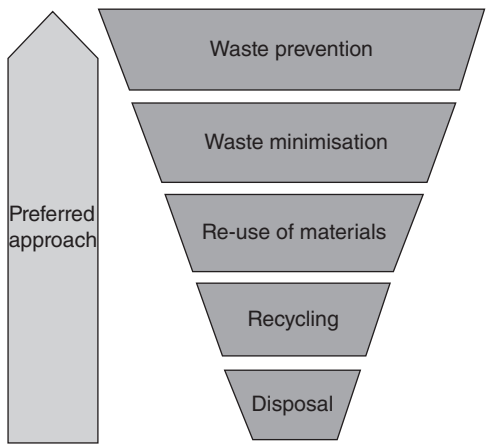
1.8 Radioactive waste management steps leading to storage and eventual disposal.

for both primary wastes from the original nuclear cycle and secondary wastes generated by reprocessing and clean-up operations. Waste minimisation programmes were largely deployed in the 1970s and 1980s. The largest volume of radioactive waste from nuclear power production is LLW. Waste minimisation programmes have achieved a remarkable tenfold decrease of LLW generation over the past 20 years, reducing LLW volumes to approximately 100 m^3 annually per 1 GW(e).

Recycling means recovery and reprocessing of waste materials for use in new products. Recycled waste can be substituted for raw materials reducing the quantities of wastes for disposal as well as potential pollution of air, water, and land resulting from mineral extraction and waste disposal. However, recycling has certain limitations when applied to radioactive materials. Due to their inherent radiation, radionuclides are much more difficult to recover from contaminated materials. Recovery usually presumes concentration of species into a smaller volume even though this may result in more dangerous materials. Waste radionuclides recovered from contaminated materials are difficult to recycle in new devices or compounds. Hence even materials which contain large amounts of radioactive constituents (e.g., SRS) often are immobilised (conditioned) and safely stored and disposed of rather than recycled.



1.9 Schematic of radioactive waste management activities.



1.10 The waste hierarchy used in the UKs decommissioning programme.

One example of recycling in the nuclear industry is of spent fuel. There are 435 currently operating NPPs in 30 countries which produce 368.2 GWe. A typical NPP generating 1 GW(e) produces annually approximately 30 t of SF. The annual discharges of spent fuel from the world's power reactors total about 10,500 tonnes of heavy metal (t HM) per year and the total amount of SF that has been discharged globally is approximately 334,500 tHM (Bychkov, 2012). During use, only a fraction of fuel is burnt, generating electricity but also forming transmutation products that may poison it. After use, the fuel elements may be placed in storage facilities with a view to permanent disposal or be reprocessed to recycle their reusable U and Pu. Most of the radionuclides generated by the production of nuclear power remain confined within the sealed fuel elements. Currently only a fraction of SF is reprocessed in countries such as France and the UK, although countries with large nuclear power programmes such as Russia and China plan to significantly increase the reprocessing capacity (Table 1.7). Also the US is reviewing the approach to open nuclear fuel cycle considering reprocessing as a viable option.

Despite the complexity of such a process, recycling of fissile elements (U, Pu) from SF results in a significant reduction of toxicity of the radioactive wastes (Fig. 1.11).

Another potential example of recycling in the nuclear industry is of military grade Pu, much of which is stockpiled in the US, Russia and the UK; a legacy of the Cold War. Since 1972, world production of plutonium has exceeded demand for all purposes. The total world plutonium inventory is not reported but a rough calculation indicates at least 2,000 metric tonnes at the beginning of the twenty-first century. It is technically possible to convert this material into a mixed U/Pu oxide (MOX) reactor fuel so that it can be used to generate energy in a suitable nuclear reactor. MOX nuclear fuel consists either of UO_2 and PuO_2 either as two phases or as a single phase solid solution $(U,Pu)O_2$ (Burakov *et al.*, 2010). The content of PuO_2 may vary from 1.5 to 25–30 wt% depending on the type of nuclear reactor. Whereas most efficient burning of plutonium in MOX can only be achieved in fast reactors, it is currently used in thermal reactors to provide energy, although the content of unburnt plutonium in spent MOX fuel remains significant (>50%).

Key aspects of waste management are to reduce the hazards associated with wastes and the volume of the waste material. Hazard can be reduced substantially by converting highly mobile liquid or gaseous wastes into stable solid forms using the techniques indicated in Figs 1.8 and 1.9. Immobilisation reduces the potential for migration or dispersion of contaminants including radionuclides. The IAEA defines immobilisation as the conversion of a waste into a waste form by solidification, embedding or

Table 1.7 Spent fuel recycling capacities, tonnes per year (Bychkov, 2012)

Country	Site	Plant (reactor type SF)	Start of operation	Capacity	
				Actual	Planned
China	Lanzou	RPP (LWR)	2008	50	50
		CRP (LWR)	2020	–	800
France	La Hague	UP2-800 (LWR)	1994	800	800
		UP3 (LWR)	1990	800	800
India	Trombay	PP (Research)	1964	60	60
	Tarapur	PREFRE1 (PHWR)	1974	100	100
	Kalpakkam	PREFRE2 (PHWR)	1998	100	100
		PREFRE3A (PHWR)	2005	150	150
	Tarapur	PREFRE3B (PHWR)	2005	150	150
Japan	Tokai-mura	PNC TRP (LWR)	1977	90	90
	Rokasho-mura	RRP (LWR)	2012	800	800
Russia	Chelyabinsk	RT1 (WWER-440)	1971	400	400
	Krasnoyarsk	RT2 (WWER-1000)	2020	–	1500
UK	Sellafield	B205 (GCR)	1967	1500	–
		THORP (LWR/AGR)	1994	900	900
Total				5,900	6,700

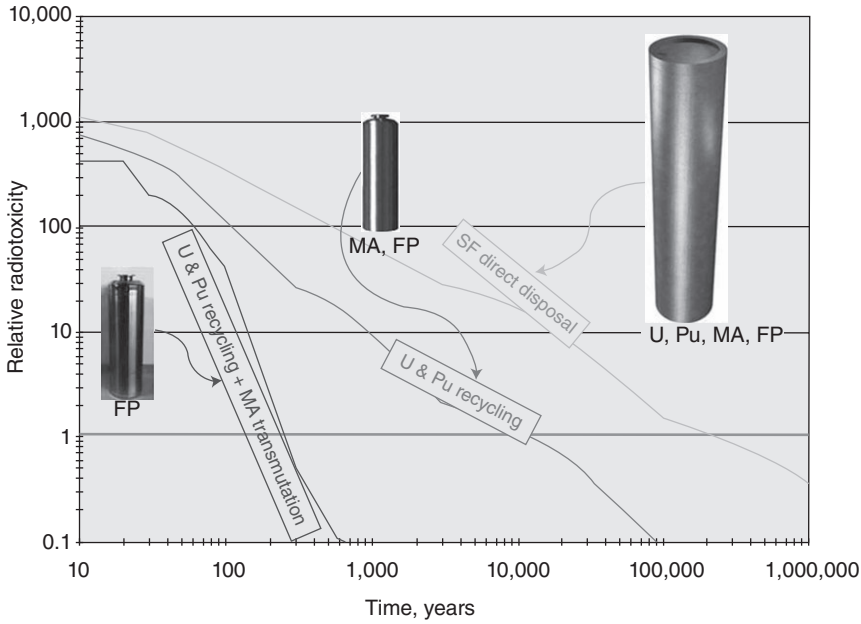
LWR: light water reactor; PHWR: pressurised heavy water reactor; WWER: water-water energy reactor; GCR: gas cooled reactor; AGR: advanced gas cooled reactor. Source: Bychkov (2012).

encapsulation. It facilitates handling, transportation, storage and disposal of RAW. Another term closely linked with immobilisation is conditioning.

Treatment of primary RAW includes operations intended to benefit safety and economy by changing the waste characteristics. Three basic treatment objectives are:

- volume reduction
- removal of radionuclides
- change of physical state and chemical composition.

As seen in Figs 1.8 and 1.9, such operations include: incineration of combustible waste or compaction of dry solid waste (volume reduction); evaporation, filtration or ion exchange of liquid waste streams (radionuclide removal); and neutralisation, precipitation or flocculation of chemical species (change of composition). The waste volume reduction factor (VRF) of a treatment process is defined as the ratio of initial volume of the treated waste V_0 to the final volume after treatment V_f: VRF = V_0/V_f. The higher the VRF, the more efficient is the treatment process. However, volume reduction inevitably leads to concentration of radionuclides which may impact on the safety and economics of the process. Treatment may lead to



1.11 Relative radiotoxicity of SF and resulting HLW on reprocessing and recycling. FP, fission products; MA, minor actinides. The time required to achieve the initial toxicity of uranium ore is significantly reduced on recycling and transmutation of MA.

several types of secondary RAW such as contaminated filters, spent resins and sludges. After treatment, depending on the radionuclide content in the waste, it may or may not require immobilisation.

1.5.3 Immobilisation options

Choosing a suitable waste form for nuclear waste immobilisation is difficult and durability is not the sole criterion. In any immobilisation process where radioactive materials are used, the process and operational conditions can become complicated, particularly if operated remotely and equipment maintenance is required. Therefore priority is given to reliable, simple, rugged technologies and equipment, which may have advantages over complex or sensitive equipment.

A variety of matrix materials and techniques is available for immobilisation (NRC, 2011). The choice of the immobilisation technology depends on the physical and chemical nature of the waste and the acceptance criteria for the long-term storage and disposal facility to which the waste will be consigned. A host of regulatory, process and product requirements has led to the investigation and adoption of a variety of matrices and technologies

for waste immobilisation. The main immobilisation technologies that are available commercially and have been demonstrated to be viable are *cementation*, *bituminisation* and *vitrification*.

Immobilisation can be simply physically surrounding the waste in a barrier material (largely the case in cementation) or chemically incorporating it into the structure of a host material (largely the case in vitrification).

Cementation uses hydraulic cements to physically surround solid ILW that is contained in steel drums (Fig. 1.12a). Ordinary Portland cement (OPC) is the most common type of cement used for immobilising liquid and wet solid wastes worldwide. Several OPC-based mixtures are currently used to improve the characteristics of waste forms and overcome the incompatibility problems associated with the chemical composition of certain types of radioactive waste. Composite cement systems may use additional powders as well as OPC such as blast furnace slag (BFS) and pulverised fuel ash (PFA). These offer cost reduction, energy saving and potentially superior long-term performance. As well as the waste form matrix, OPCs will be used in structural components of any GDF (such as walls and floors) and are potential backfill materials, so an understanding of their durability in an underground environment even without waste is important.

(a) (b)

1.12 (a) 500 litre steel drums containing cemented ILW and (b) vitrified waste in 2 m tall steel containers.

Embedding radioactive waste in bitumen has been used in immobilisation since the 1960s and the total volume of RAW immobilised in bitumen currently exceeds 200,000 m^3. In the bituminisation process, radioactive wastes are embedded in molten bitumen and encapsulated when the bitumen cools. Bituminisation combines heated bitumen and a concentrate of the waste material in either a heated thin film evaporator or extruder containing screws that mix the bitumen and waste. The waste is usually in the form of a slurry, for example salt aqueous concentrates or wet ion exchange resins. Water is evaporated from the mixture to about 0.5% moisture, intermixed with bitumen so that the final product is a homogeneous mixture of solids and bitumen, termed bitumen compound. Its retention properties usually exceed those of cements at higher waste loadings. Bituminisation is particularly suitable for water-soluble RAW such as bottom residues from evaporation treatment and spent organic ion exchangers. However, a drawback of bitumen is its potential fire hazard. The possibility of combustion in the case of an accidental fire has led to certain restrictions on the use of bitumen as an immobilising matrix.

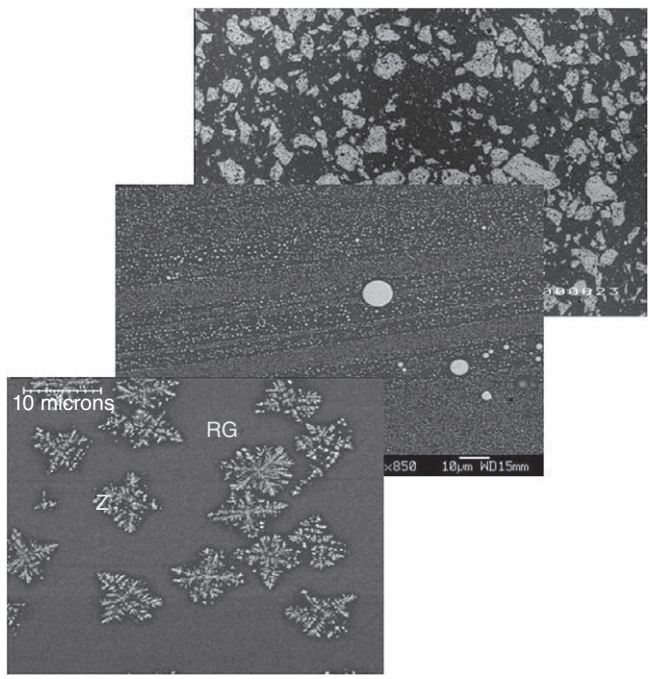
Vitrification is an attractive immobilisation technique because of the small volume of the resulting waste form (Fig. 1.12b), the large number of elements which can be incorporated in it and its high durability. The high chemical resistance of glass allows it to remain stable in corrosive environments for long periods. Waste vitrification technology is a compromise between the desired durability of the final waste form and its processing efficiency (Ojovan and Lee, 2007). The most durable materials would require very high processing temperatures (>1500°C) which cannot be used because at high temperatures waste radionuclides occur in volatile species, generating large amounts of secondary wastes and diminishing the immobilisation efficiency. The most common glasses used in vitrification of nuclear waste are borosilicates and phosphates. Vitrification has been used for nuclear waste immobilisation for more than 40 years in France, Germany, Belgium, Russia, UK, Japan and the US. The total production of all vitrification plants by the end of 2000 was approximately 10,000 tonnes of radioactive glass in roughly 20,000 canisters. Vitrification is also currently used for immobilisation of low and intermediate level waste (LILW).

The highest degree of volume reduction and safety is achieved through vitrification, although this is the most complex and expensive method, requiring a relatively high initial capital investment. However, difficult legacy waste streams are known for which current technology is inadequate, so that new approaches must be developed. These comprise development of new waste forms such as crystalline ceramic and composite radionuclide hosts as well as of new immobilising technologies such as thermochemical and *in-situ* methods. New approaches aim also to create geochemically

stable materials in equilibrium with the disposal environment to ensure a safer nuclear waste disposal scenario.

Glass composite materials (GCMs) are used to immobilise glass-immiscible waste components such as sulphates, chlorides, molybdates and refractory materials requiring unacceptably high melting temperatures. GCMs comprise both vitreous and crystalline components (Lee *et al.*, 2006). Depending on the intended application, the major component may be a crystalline phase with a vitreous phase acting as a bonding agent, or, alternatively, the vitreous phase may be the major component, with particles of a crystalline phase dispersed in the glass matrix (see Fig. 1.13).

GCMs may be produced by dispersing both melted materials and fine crystalline particles in a glass melt and may be used to immobilise long-lived radionuclides (such as actinide species) by incorporating them into the more durable crystalline phases, whereas the short-lived radionuclides may be accommodated in the less durable vitreous phase. GCMs may also be glass ceramics where a glass is crystallised in a separate heat treatment step (Caurant *et al.*, 2009), The French have developed a U-Mo GCM to immobilise Mo-rich HLW. Another example is the GCM developed to immobilise sulphur-enriched waste streams in Russia, containing



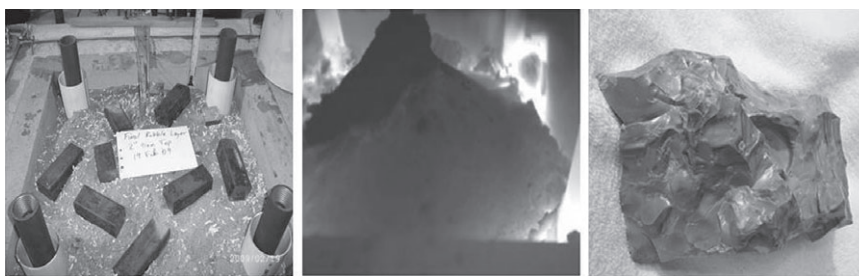
1.13 Examples of GCM microstructures.

conventional borosilicate glass vitreous phase with uniformly distributed particles comprising up to 15% by volume of yellow phase.

GCMs are being developed in many countries to immobilise their difficult wastes. For example, the UK's most hazardous wastes are those in the legacy ponds and silos (LP&S) at Sellafield. A number of novel thermal technologies are being examined to immobilise the complex, often ill-defined and heterogeneous wastes found in the LP&S. These include pyrolysis steam reforming, plasma vitrification and Joule heating in container melting (JHCM). In the latter process, mixed solids and sludge wastes are placed in a concrete lined steel container with embedded graphite electrodes in the corner (Fig. 1.14) and melted to produce a stable solid.

While JHCM can successfully convert reactive material (e.g., metals, sludges and organics) to more stable forms, the variable nature of the wastes makes control of process and product difficult, and it is difficult to characterise both the heterogeneous waste and product. Much R&D is needed including durability testing of the products of these technologies. However, their use has seriously reduced the hazard from the original wastes and pragmatic engineering approaches such as these are needed even if the resulting waste form is not as perfect as ultimately desirable.

Single-phase ceramics such as zircon ($ZrSiO_4$) can potentially host a large number of nuclides and can be used as a monophasic waste form. However, monophase ceramics are difficult to fabricate and polyphase compositions are more common. The composition of the polyphase ceramic can host multiple radionuclides and be tailored to that of the waste composition to achieve complete and reliable immobilisation of the waste constituents. The most famous polyphase ceramic for nuclear waste immobilisation is Synroc. Synroc is short for 'Synthetic Rock', invented in 1978 by T. Ringwood of the Australian National University. Synroc is made of geochemically stable natural titanate minerals which have immobilised uranium and thorium for billions of years. U/Th-containing natural analogues of the basic constituent



(a) Before (b) During (c) After

1.14 (a) In-container setup, (b) during heating, (c) resulting stable solid product.

of Synroc – zirconolites from Sri Lanka dating back 550 million years while amorphised – have nonetheless withstood the alteration processes of their natural environment.

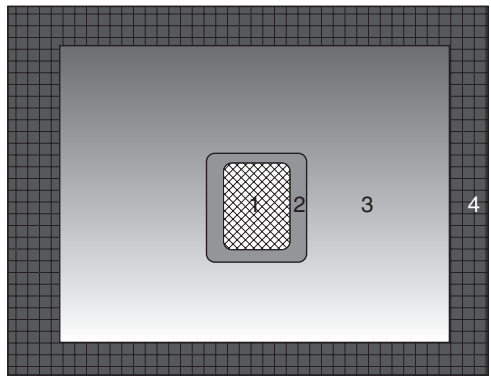
1.5.4 Storage options

When examining storage options for RAW, it is important to consider the whole storage system rather than concentrating on just the store building itself (CoRWM, 2009). A number of interacting components and operations combine and contribute to create the necessary robust, safe and secure storage arrangements. These factors must be considered in an integrated manner. There are two main concepts in the storage of RAW. If the packaged waste forms are basic, then a high quality often shielded store will be needed. On the other hand, if the waste form is high quality and shielded, then the store can be of poorer quality or the waste containers can simply be left in the open. A generic shielded store is shown in Plate I (between pages 448 and 449) and an example of a high quality store has recently been constructed at Hunterston in Scotland (Fig. 1.15(a)) which has 2 m thick reinforced concrete walls and roof and careful control of atmosphere. Figure 1.15(b) also shows the stillages containing 4 ILW drums that will be stacked on top of each other in the store (and eventually in the GDF).

The waste form or product, its container, the building structure, the ventilation system, the handling equipment, the monitoring and inspection regime and the maintenance and refurbishment regime all have roles to play in ensuring safety and security of the store. As illustrated in Fig. 1.16, the waste storage system involves a number of levels. The wasteform (1) is the primary protective barrier, the waste container (2) is the secondary barrier. Control of store environment (3) is important in maintaining the

(a) (b)

1.15 (a) Inside the store at Hunterston in Scotland, (b) stillage containing 4 ILW drums.



1.16 The four parts of the storage system (from NDA, 2012b).

integrity of the waste form and waste container, while the store structure (4) is the final layer of weather/atmosphere protection for waste package and an important element of physical security of waste.

Packages inevitably evolve during storage, and those changes affecting the safety function need to be understood and controlled to satisfy the regulators of the safety of the store and waste. Different storage concepts and designs require different performances from these various components and operations and therefore place different degrees of reliance on them. Quite different combinations of them can provide equally safe and secure storage. For example, most existing modern stores in the UK have massive concrete structures holding unshielded containers, but the alternative 'mini-store' concepts rely on heavily shielded containers within lightly built stores. This latter concept is used in some other EU countries. In a storage system, not every component need last for the whole design life. It is possible at the design stage to plan to replace or refurbish various components and build in at the outset specific features to enable this. More straightforward items to consider are building fabrics, external ventilation systems and power supplies. The more complex refurbishments or replacements to consider are cranes, active area surveillance equipment and major building structures.

In the late 1970s and early 1980s, the need for alternative storage in the US began to grow when the storage ponds at many nuclear reactors began to fill up with stored spent fuel. As there was not a national storage facility in operation, utilities began looking at options for storing spent fuel. Dry cask storage was one of the most practical options for temporary storage. The first dry storage installation in the US was licensed by the Nuclear Regulatory Commission (NRC) in 1986 at the Surry NPP in Virginia. Spent fuel is currently stored in dry cask systems at a growing number of power plant sites. The NRC estimates that the SF ponds at many US NPP will be

full by 2015, so requiring the use of temporary storage. The 2008 NRC guidelines call for fuels to have spent at least five years in a storage pool before being moved to dry casks. Due to the demise of the Yucca Mountain project, more US SF and waste is being stored in sealed metal casks filled with inert gas. Examples of high quality containers include CASTOR, which is an acronym for CAsk for Storage and Transport of Radioactive material (Fig. 1.17).

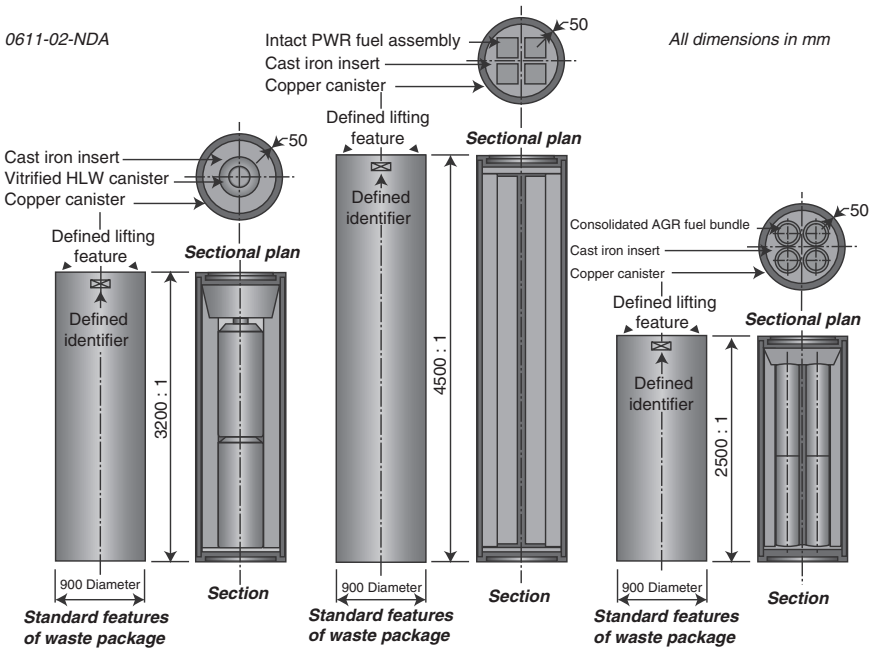
In the UK, options being examined for SF include multi-purpose containers (MPC) suitable for storage, transport and disposal of a range of SF types (Fig. 1.18). As well as high quality packages for SF, they have also been developed for less active wastes. So-called yellow boxes (Fig. 1.19) have been used extensively in Europe and used to store spent resin waste from existing storage tanks at the Dungeness plant in England. The containers are transportable and offer self-shielded protection, weighing around 18 tonnes when empty. The waste is expected to be stored in them for at least a decade.

As more waste is generated and being stored, but in many countries without an end-point of geological disposal in sight, an issue is whether to store all waste at sites or to consolidate wastes at centralised national or regional stores. The BRC, for example, recommended this option be examined in the US and it is also being considered in the UK.

1.5.5 Disposal options

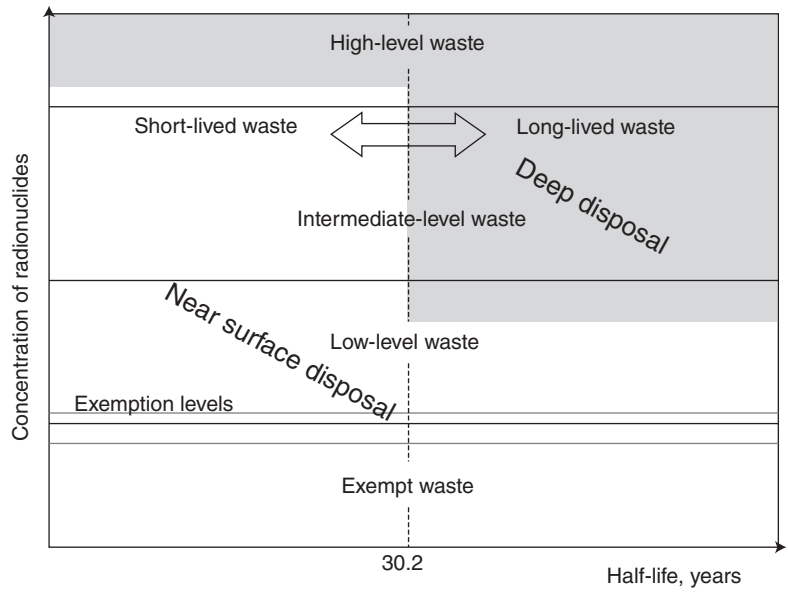
Options for disposal are indicated in Fig. 1.20 and depend to large extent on the content and half-life of radionuclides in the waste. Small contents and short-lived wastes may be suitable for near surface disposal (IAEA, 2002), while larger contents and long-lived radionuclides require deep or

1.17 Open air CASTOR store for US SF.



1.18 MPC options under development for UK SF.

1.19 Yellow boxes for storing ILW.

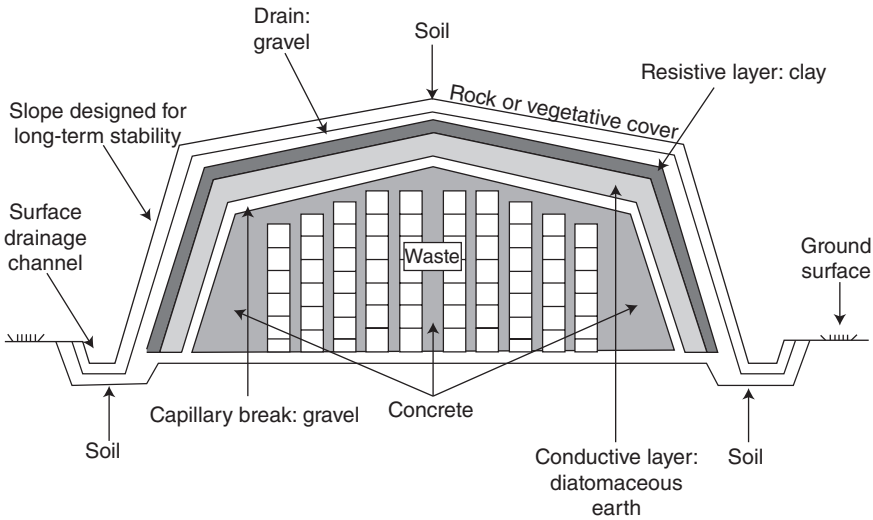


1.20 Illustration of disposal and storage options for types of radioactive wastes.

very deep disposal, relying on the geosphere to keep the radioactive species from the biosphere (IAEA, 2003a; Ahn and Apted, 2010). As for the storage concepts described above, most geological approaches use a multi-barrier system to improve the safety of disposal where the waste form, container, near field environment (e.g., engineered barrier system, EBS) and far field environment (host rock) are all important in retaining radionuclides in the geosphere.

Near-surface disposal

Near-surface disposal sites are constructed anywhere from on the surface itself to up to 60 m below it. Such facilities with an EBS are suitable for most LLW and LILW and are widespread across Europe (e.g., Drigg in the UK, El Cabril in Spain) and are also used in the US and Japan. Globally we have decades of experience of operating such disposal sites. The EBS, which typically consist of clay or other barrier layers, is necessary to reduce the leach rate of radionuclides from the waste and to divert water away from the wastes. Water management is used during the operational phase of these facilities when the waste packages are uncovered as water cannot be allowed to accumulate within the waste cells. A typical system is shown schematically in Fig. 1.21. Near-surface storage facilities are appropriate where wastes contain small amounts of short-lived wastes.



1.21 Schematic of a surface repository.

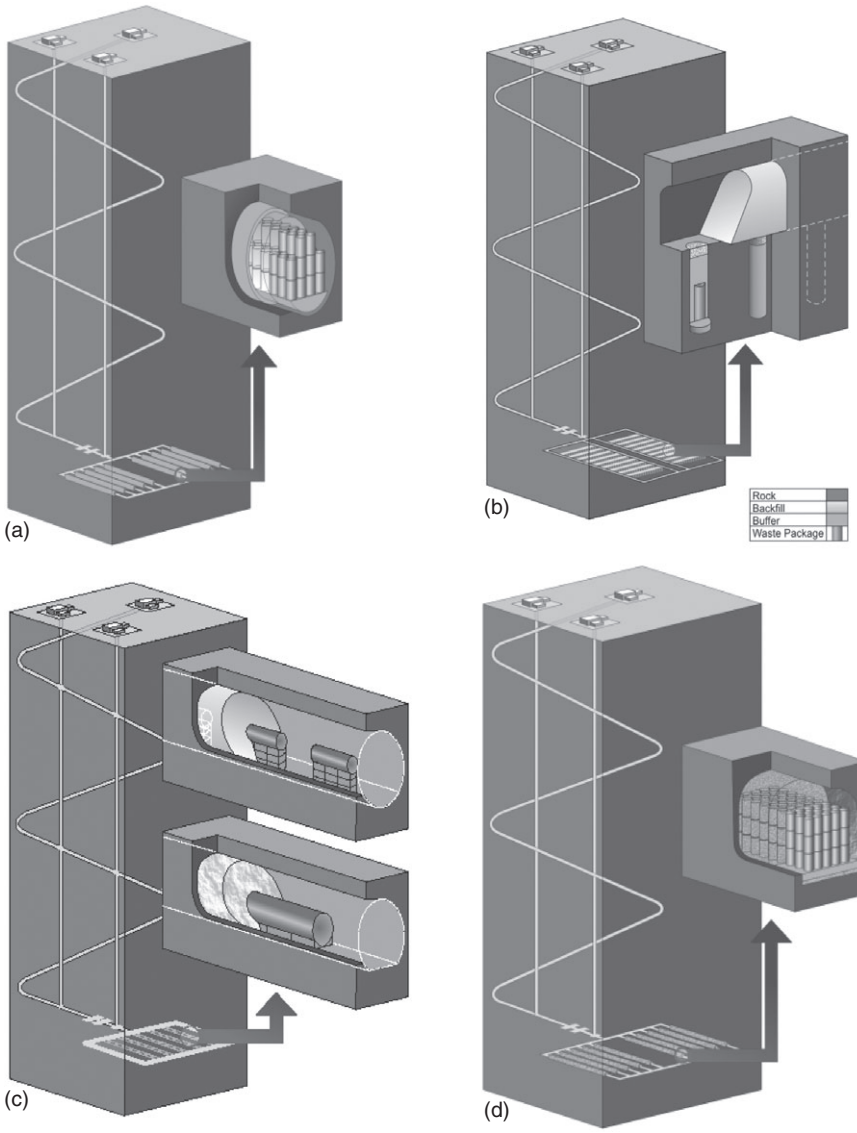
Deep geological disposal

Geological disposal in a mined repository is the most likely option for HLW, SF, SRS and long-lived LILW (IAEA, 2003a; Ahn and Apted, 2010). The main concepts of geological disposal are wet and dry, typically at depths from 500 to 1,500 m.

The wet option is a mined and engineered repository located so that eventual water ingress and saturation is inevitable. Various types of host rock are being considered governed largely by the local geology including hard rock (e.g., granite as in the Swedish and Chinese concepts) and soft rock (e.g., clays in France and Belgium).

The dry mined and engineered repository concept was favoured in the US, including high and dry (Yucca Mountain, Nevada) and shallow and dry (the Waste Isolation Pilot Plant (WIPP) located in salt in Carlsbad, New Mexico. However, technical (and other) problems at Yucca Mountain, including that it was not as dry as hoped, have led to the demise of that programme.

Figure 1.22 shows a range of GDF concepts (NDA, 2010b). The concept that will be chosen for the GDF is dependent on rock type, volume of rock available and wastes. Various combinations of tunnel, backfill and waste may be appropriate and different wastes may be kept in different parts of the repository. In the UK, a range of options are being considered while the MRWS process progresses. Until a site and geology is selected, these remain concepts but they do allow initial work on safety cases to start. For example, the concept in Fig. 1.22(a) would be used for ILW in vaults in

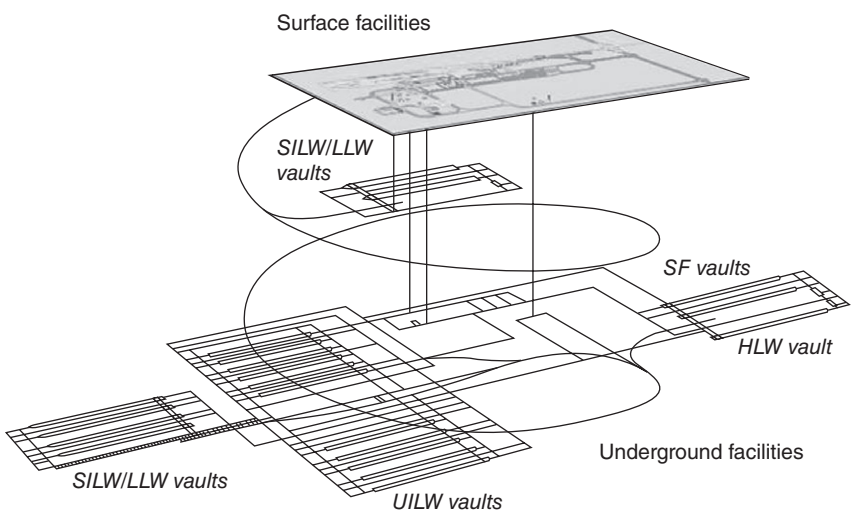


1.22 (a–d) A range of disposal options for a mined GDF being considered in the UK.

lower strength rock with cement grout backfill, while that in Fig. 1.22(b) is appropriate for SF in vertical boreholes in high strength rock (granite) with clay buffer and backfill. The concept in Fig. 1.22(c) is for SF in horizontal tunnels in waste containers in a low strength sedimentary rock (clay) with clay buffer and that in Fig. 1.22(d) covers ILW in vaults in higher strength rock with a cement grout backfill.

The UK concept is for multiple vaults in the same region (Fig. 1.23) to accommodate the complex array of waste forms that we have, a legacy of our early indecision on which reactor type to build and of military and research programmes. A clear research need is to understand waste form evolution during storage and disposal and the eventual interaction of the corrosion products from the different parts of the GDF.

In 1980, the IAEA-sponsored International Nuclear Fuel Cycle Evaluation (INFCE) waste management and disposal report recommended that proposals 'for establishing multinational and international repositories should be elaborated' due to their non-proliferation advantages. In 2003, Mohamed El Baradei, Director-General of the IAEA, said to the UN General Assembly: 'We should … consider multinational approaches to the management and disposal of spent fuel and radioactive waste. Over 50 countries currently have spent fuel stored in temporary locations, awaiting reprocessing or disposal. Not all countries have the appropriate geological conditions for such disposal – and, for many countries with small nuclear programs, the financial and human resources required for the construction and operation of a geological disposal facility are daunting. … Centralised facilities for disposal of spent fuel and/or vitrified high-level wastes would reduce the diversion risk and be more economical.' More recently, in 2011, the Council of the European Union adopted a Directive on the responsible and safe management of spent fuel and radioactive waste, highlighting that each member state has ultimate responsibility for management of the spent fuel and waste generated in its territory. However, the door was left open for small countries with limited volumes of waste to share regional



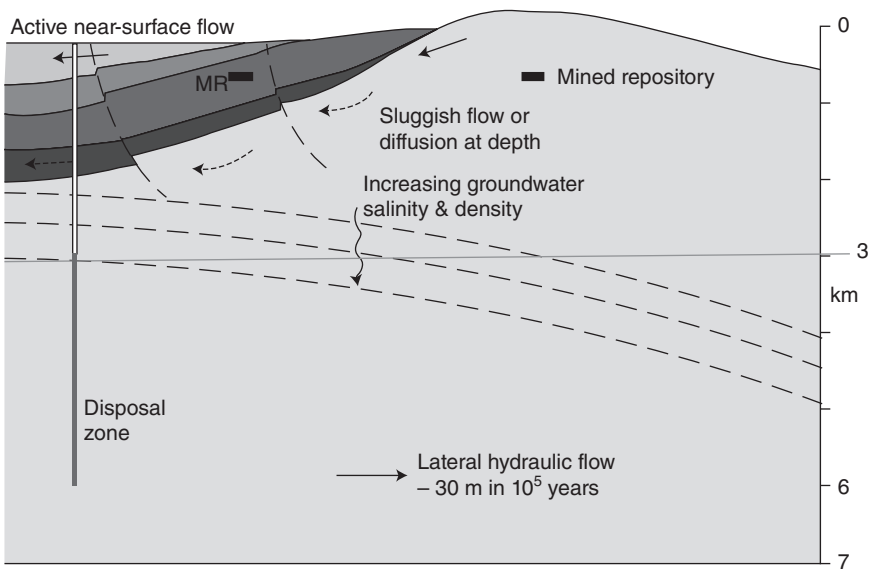
1.23 The UK's current multi-vault GDF concept.

repositories. The main message is that the option of EU Member States sharing repositories is included by Clause 3 in Article 4 on General Principles which states that '*Radioactive waste shall be disposed of in the Member State in which it was generated, unless at the time of shipment an agreement … has entered into force between the Member State concerned and another Member State or a third country to use a disposal facility in one of them.*' This implies that regional cooperation could be an important aspect of the detailed plans that the EC expects Member States to produce within four years. Austria, Bulgaria, Ireland, Italy, Lithuania, Netherlands, Poland, Romania, Slovakia and Slovenia, have resolved to move towards setting up a European Repository Development Organisation (ERDO).

Very deep geological disposal

Another concept is that of very deep (permanent) disposal. In this concept (Gibb, 2000), the waste is located at depths of 3 km or more and as such any transport of radionuclides through the geosphere is extremely limited (Fig. 1.24). Further, if located in suitable (granitic) rock, the radiogenic heat from HLW can cause reaction with the surrounding rock and lead to creation of a sarcophagus or granite coffin which seals in the waste permanently.

The US BRC was positive about the deep borehole disposal concept and the US is planning a demonstration programme. However, this is untried



1.24 Schematic of very deep borehole disposal.

technology that requires a case to be made for its safety that potentially could take many decades to come to fruition.

1.6 Strategies for managing uncontrolled releases and contaminated site clean-up

Managing uncontrolled releases and contaminated site clean-up are considered in detail in Chapter 8 and Part III so only a summary derived from them is provided here. Agreed international safety requirements cover such situations (IAEA, 2003c). To ensure that protective measures can be quickly and efficiently implemented to mitigate the adverse effects of an accident or other forms of long-term contamination at a nuclear site requires good planning, clear strategies and a good managerial team. Preparations for environmental remediation should ideally be done in two phases: preliminary planning, which should be available as part of normal operation, or emergency preparedness for each nuclear facility; and detailed remediation planning, which takes into account site (and accident where applicable) specific information.

According to the IAEA Safety Glossary (IAEA, 2007) remediation is defined as any measure that may be carried out to reduce the radiation exposure from existing contamination of land areas through actions applied to the contamination itself (the source) or to the exposure pathways to humans. An important element in the overall remediation concept is that complete removal of the contamination is not implied. Remediation aims to achieve optimised protection of the public, workers and the environment. The goal of remediation activities is the timely and progressive reduction of hazard and eventually, if possible, unrestricted release of the site. However, there are situations where this goal may not be achievable and then it must be demonstrated that as a minimum any unacceptable risks to human health and the environment have been removed. When choosing a remediation option, a range of factors must be considered, such as the impacts on health, safety and the environment; and technical, social and financial factors. National remediation strategies are needed to specify, prioritise and to ensure remediation situations are managed in a manner commensurate with the risks associated with the contaminated areas and taking into account possible effects on neighbouring countries.

In general, remediation of a contaminated area involves preparation and approval of a remediation plan; remediation operations; and management of RAW resulting from the remediation activities. It needs to be based on collection and assessment of all available information of current and past activities at the site. Therefore an appropriate assessment of both the radiological and non-radiological impacts of the situation must be performed and the benefits and detriments associated with possible remedial measures,

including the associated restrictions and institutional arrangements following remediation must be identified based on established reference levels as part of the decision-making process.

The remediation plan has to be subject to the approval of the regulatory body prior to its implementation and must state, as a minimum: the goal for the remediation; reference levels for remediation; the nature, scale and duration of the remedial measures to be implemented; the waste disposal or storage site, as appropriate; any post-remediation restrictions; and the monitoring and surveillance programmes and arrangements for institutional control for the remediation area. During the implementation of remedial measures, consideration must be given to (i) radiation safety, transport safety and waste safety, general health and safety issues and environmental issues so as to minimise hazardous impacts, and (ii) the potential for prolonged exposure after the termination of remediation activities.

The area has to be monitored and surveyed regularly during remediation so as to verify the levels of contamination; to ensure compliance with the requirements for site release and for waste management, and to detect any unexpected levels of radiation. Before an area can be released for unrestricted use, a survey must be performed to demonstrate that the end-point criteria and conditions, as established by the regulatory body, have been met. The organisation responsible for the surveillance and verification of activities must be clearly identified. An appropriate programme, including any necessary provisions for monitoring and surveillance, has to be established to verify the long-term effectiveness of the completed remedial measures. As part of the overall management system, arrangements for archiving, retrieval and amendment of all important records concerning the initial characterisation of the area, the choice of options for remediation and the implementation of remedial measures, including all restrictions and the results of all monitoring and surveillance programmes, must be established and maintained in all cases.

1.7 Sources of further information

UK websites

- Department of Energy and Climate Change (DECC) Managing Radioactive Waste Safely (MRWS) programme (www.mrws.decc.gov.uk),
- NDA (www.nda.gov.uk).
- CoRWM (www.corwm.decc.gov.uk).

International websites

- IAEA (www.iaea.org),
- WNA (www.world-nuclear.org).

Useful information can be found in the following textbooks

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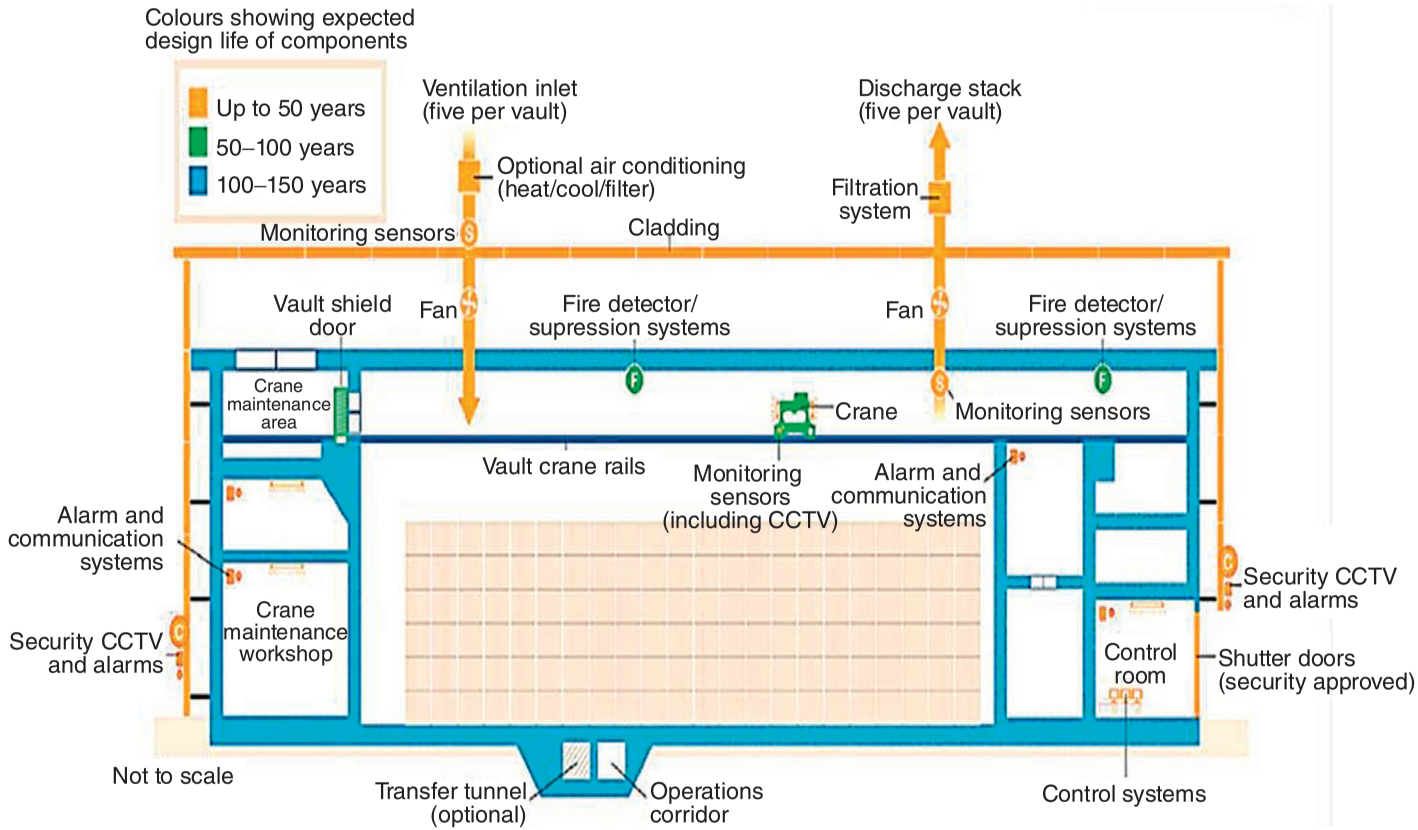

Plate I (Chapter 1) Generic shielded store (from NDA, 2012b).

13

Nordic countries: experience of radioactive waste (RAW) management and contaminated site clean-up

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Abstract: The chapter describes the historical background to the current radioactive waste (RAW) situation in the Nordic countries. It discusses the current management and final disposal of low level waste (LLW) and intermediate level waste (ILW) and the siting processes for a repository for spent nuclear fuel. Early nuclear activities in Sweden led to contaminated nuclear facilities and uranium mining sites. The chapter describes the ongoing remediation of these sites.

Key words: nuclear waste, repository siting, uranium mining, site clean-up, legal framework.

13.1 Introduction

The atomic bombs dropped over Hiroshima and Nagasaki alerted Sweden to the potential of nuclear energy. Until then, the programmes for nuclear physics research had been very limited. In the autumn of 1945, however, the Swedish Defense Research Establishment (FOA) asked for funding for preliminary studies. The military thought that it would be useful for a small country to possess an atomic bomb as a deterrent (Jonter, 2002). The peaceful aspects of nuclear energy were, however, most important. In 1945 a committee, Atomkommittén, was formed. Its task was to plan future nuclear research and to find applications for peaceful use of nuclear energy. The committee came to the conclusion that the government should develop this new power source in cooperation with industry and in 1947 AB Atomenergi was constituted with the government as the main shareholder (Larsson, 1987; Elam and Sundqvist, 2006).

Much of the initial research was concentrated on producing uranium and separating plutonium from irradiated uranium. The idea was that Sweden should become independent and self-sufficient in energy supply. The uranium was to be mined from the shale deposits in south Sweden. Any import of uranium was at that time out of the question. With a limited supply of uranium, the solution was a heavy water reactor with natural or low enriched uranium. The heavy water was to be imported from Norway.

The first Swedish reactor, R1, put into operation in July 1954, was constructed underground at the campus of the Royal Institute of Technology (KTH) in central Stockholm. It was fuelled, however, with uranium borrowed from France. The agreement was that Sweden would return the uranium as soon as the Swedish uranium mines had gone into production.

R1 was a research reactor intended neither for energy production nor for plutonium production. Therefore, a second step was planned. In a sparsely populated coastal area with access to water, one or more reactors were to be built. The final location was Studsvik, where AB Atomenergi built its research centre. This was also the location of R2, a materials testing reactor, which was started in 1961 (see Fig. 13.1).

During the latter part of the 1950s, following a conference in Geneva, the nuclear weapons countries made available on the market enriched and natural uranium. The Swedish government issued a nuclear energy law in 1956, which allowed the development of nuclear power. This boosted the Swedish national nuclear programme and AB Atomenergi proposed the construction of two more reactors, R3 and R4. A group of private power companies had already in 1955 formed a consortium, Atomkraftkonsortiet (AKK). Their purpose was to follow the international development, propose reactor types and finally build a nuclear power plant (NPP) for the owners. AKK was first to propose light water reactors in Sweden.

The government policy was, however, still heavy water reactors and domestic supply of uranium. The programme was very optimistic, but it soon became obvious that the country did not have the means to carry it out. Of the originally foreseen five to six heavy water reactors built before 1965, only one was built and started in 1963 in Ågesta in southern Stockholm. This reactor was mainly used for district heating and operated until 1973. While Sweden concentrated on the heavy water line, light water reactors were developed in the US. The development of boiling water reactors (BWR) and pressurized water reactors (PWR) was rapid, while the Swedish national nuclear programme ran into difficulties. The programme included uranium production, fuel factories and reprocessing facilities. In 1965 the uranium production facility in Ranstad was opened. At the time, however, the cost for uranium from Ranstad was considerably higher than the world market price. Mining stopped in 1969 and the facility was closed in the early 1970s.

Vattenfall and AB Atomenergi had cooperated in building the Ågesta reactor and were now planning a larger reactor in Marviken. The reactor design was changed several times and finally it was decided it should have a power of 400 MW and also be used for plutonium production. At the same time, AKK decided in 1959 to build a small BWR north of Oskarshamn. In 1965 AKK was transformed into Oskarshamns Kraftgrupp AB (OKG), and a BWR reactor was finally ordered in 1966 and in

13.1 Map showing sites of nuclear installation activities in Sweden and Finland.

operation in 1973. Following the order for the first reactor at Oskarshamn, 11 more reactors were ordered and put into operation during the following two decades.

The Marviken reactor was ready for test operation by 1968. That year Sweden had signed the non-proliferation treaty and there was no longer any reason for plutonium production. The result of a government investigation published in 1968 led to the formation of ASEA-ATOM (1969), owned

half each by ASEA[1] and the state. The reactor design and nuclear fuel activities were transferred to this new company. AB Atomenergi continued as a research institute. This marked the end of the Swedish national nuclear programme. The Marviken reactor was never started. The legacy of the programme, however, was a uranium production facility and a pilot facility for reprocessing of nuclear fuel.

During the 1970s, the use of nuclear energy became increasingly controversial. In 1977, a new law required that the nuclear industry demonstrate how the nuclear waste was to be taken care of before any reactor could be fuelled. This led to the launching of the project Kärnbränslesäkerhet (KBS). The project finally resulted in the fuelling of all the reactors finalized after the law became effective. In 1981 a new law required that the nuclear power companies fund the future costs of nuclear waste management. The industry delegated a company, Swedish Nuclear Fuel Supply Company (SKBF, founded in 1973), jointly owned by Sydkraft AB (now E.ON Kärnkraft Sverige AB), Vattenfall AB, OKG Aktiebolag and Forsmarks Kraftgrupp AB, to perform the necessary research and development work. The company name was later changed to the Swedish Nuclear Fuel and Waste Management Company (SKB).

At this time, Sweden has in operation three BWR in Oskarshamn, three BWR in Forsmark and two BWR and two PWR in Ringhals. The two reactors in Barsebäck were closed in 1999 and 2005, respectively.

The Finnish situation was different from that in Sweden. Finland had been on the losing side in the Second World War and had to cede 10% of its territory to the Soviet Union and was, furthermore, obliged to pay 300 million dollars in war reparations to the Soviet Union, following the 1947 Paris Peace Treaty. The loss of Karelia also meant the loss of important hydropower plants. After the war, Finland only had about two-thirds of its hydropower left compared to the situation before the war. Finland lacked both energy and economic resources to embark on a nuclear research programme. Furthermore, the Paris Peace Treaty prohibited Finland from research and development of nuclear weapons materials. That, and the lack of resources, meant that Finland did not invest in a research reactor as early as the other Nordic countries. The expanding Finnish industry, however, needed electricity and the potential of nuclear energy for electric power generation was recognized early (Anttila, 2000; Kojo, 2006).

After President Eisenhower launched the 'Atoms for Peace' initiative and the possibilities of having access to nuclear energy without a costly

[1]ASEA (Allmänna Svenska Elektriska Aktiebolaget) bought the state's share of the company in 1982. After ASEA merged with the Swiss Brown Boveri Corporation forming ABB (ASEA Brown Boveri), the name was changed to ABB Atom. Since 2000, the company is part of the Westinghouse Electric Company as Westinghouse Electric Sweden (2003).

domestic development programme, the Finnish Academy of Science suggested the formation of an Atomic Energy Committee in 1954, which the Finnish government then set up in 1955. The tasks of the committee included investigating the suitability of nuclear energy in Finland. Parallel to this, the industry established a company, Atomienergia Oy (Atomic Energy Ltd), primarily to satisfy the interests of the forestry industry. A milestone in Finnish nuclear research was taking into operation a TRIGA-type research reactor in 1962. This reactor is still in operation.

In 1965, the state-owned energy company, Imatran Voima (IVO, now Fortum Power and Heat Oy, FPH) put out to tender for a nuclear power plant to different suppliers, and in 1969 IVO decided to buy from the Soviet Union. Two PWR-type reactors were ordered and put into operation in 1977 and 1980 at Hästholmen near Loviisa. In 1969, the private industrial companies formed Teollisuuden Voima Oy (TVO) and the following year TVO decided to build two reactors. These were BWR reactors from ASEA ATOM and they were taken into operation in 1978 and 1980 in Olkiluoto. At present, TVO is having a third reactor constructed. Finland is, therefore, the only Nordic country expanding its nuclear energy capacity with new reactors.

There is no commercial electricity-generating reactor in Norway. In fact, over 99% of all electricity in Norway is produced by hydropower (OECD-NEA, 2005). Norway has, however, been very active in nuclear research. This started immediately after the Second World War, first at the Norwegian Defense Research Institute (FFI) from 1946 and later at the Institute for Nuclear Energy (IFA, now Institute for Energy Technology, IFE), which was founded in Kjeller in 1948. Norway's and also Scandinavia's first nuclear reactor, JEEP, was started at IFA as early as in 1951 (Oberländer *et al*., 2009; OECD-NEA, 2005). In all, there have been three research reactors at IFA, JEEP (1951–1967), NORA (1961–1967) and JEEP II (1967–). In addition to these, there is a fourth research reactor, the Halden boiling water reactor (HBWR) in Halden.

As was the case for Norway, there is no commercial electricity-generating reactor in Denmark. Three research reactors, DR 1, DR 2 and DR 3, have been operated at the Risø National Laboratory. They were started between 1957 and 1960 and are now all shut down, and DR 1 and DR 2 are fully decommissioned (Dansk Dekommissionering, 2006, 2009).

13.2 Sources, types and classification of wastes

The commercial electric power reactors in Finland and Sweden generate by far the majority of the radioactive waste (RAW) in the Nordic countries. The waste is classified into three categories: operational waste or reactor waste, decommissioning waste and spent nuclear fuel.

The treatment of the waste depends on the activity level. The operational waste, which accounts for about 85% of all wastes from the reactor operations, consists mostly of low and intermediate level waste that requires isolation from the environment for at least 500 years. The low level waste (LLW) can generally be handled without radiation shielding, while some shielding will be necessary for the intermediate level waste (ILW).

The decommissioning waste is mostly the scrap metal and concrete from the future dismantling of the reactors. Most of the waste will be LLW and ILW, but the reactor pressure vessel and its internal components are classified as long-lived waste and must be isolated for thousands of years.

The spent nuclear fuel is only a small fraction of the waste, but it is the most dangerous waste that must be isolated for very long time periods.

In addition to these major waste sources, there is also waste from research and industrial and medical applications. This will also include some research reactor fuel. For some research reactor, e.g., Studsvik and Risø, however, the fuels were on loan from the US and have been shipped back there.

13.3 Radioactive waste (RAW) management strategies: history and developments

Throughout the 1960s and during the early part of the 1970s, Sweden saw reprocessing as the preferred route for spent fuel management. Research and development work at Studsvik aimed at this, and as late as 1976, a government investigation proposed a reprocessing facility to be built in Sweden. In line with that policy, in 1969 OKG signed a reprocessing contract with UKAEA (now a site license company of the UK Nuclear Decommissioning Authority, NDA) for the reprocessing of 140 tonnes of nuclear fuel. The fuel was shipped to Sellafield between 1974 and 1982; however, it was not reprocessed until 1997. No high level waste will be returned to Sweden, but the plutonium content will be used in mixed oxide (MOX) fuel (Persson, 1992; Wikdahl, 2005).

In 1976, the new Swedish government required in its policy statement that an acceptable contract for reprocessing of the nuclear fuel and a safe method for disposal of the RAW before the second reactor at Barsebäck could be fuelled and put into operation. As a consequence of this, Sydkraft, the owner of Barsebäck, signed a contract with Cogéma for reprocessing of 57 tonnes of fuel for the remainder of the 1970s. In 1978, contracts were signed for reprocessing of 672 tonnes to obtain the licenses to operate reactors in Forsmark and Ringhals and for continued operation of Barsebäck 2. To meet the requirement to demonstrate a method for safe disposal of the nuclear waste, the Swedish nuclear power industry started project Kärnbränslesäkerhet, KBS (Nuclear Fuel Safety) and published the first

report, commonly referred to as KBS-1 in 1977 (KBS, 1978). In 1982 a special ship, M/S Sigyn was launched for transporting spent fuel to France for reprocessing and to the joint intermediate fuel storage facility, Clab, the construction of which started in 1980 and was commissioned in 1985. The facility is 30 meters underground and has two storage pools with a capacity of 8,000 tonnes.

When later the responsibility of the reactor owners for managing the nuclear waste was formalized in the 'Stipulation Act' (*villkorslagen*), the possibility for direct disposal of the spent nuclear fuel had been included. The reprocessing capacity was very limited and it was necessary to also demonstrate safe disposal of spent nuclear fuel. Immediately after KBS-1, work started on preparing a preliminary safety analysis of a repository for spent nuclear fuel, KBS-2 (KBS, 1979). This analysis was submitted to international review but not submitted to the Swedish authorities. Instead it was further elaborated and finally submitted as KBS-3 in order to have direct disposal of spent nuclear fuel accepted as a waste management option (KBS, 1983). The safety analysis was published in 1983. In 1984, the Stipulation Act was incorporated in the new Nuclear Activities Act. Based on KBS-3, the remaining two reactors, Forsmark 3 and Oskarshamn 3, were licensed and fuelled. This new law also required that the nuclear power industry submit a research programme for the next six years and that such a programme was to be submitted every three years for review and approval by the authorities and the government. The nuclear power companies delegated the research and development work to their jointly owned company SKBF, which from 1984 was renamed the Swedish Nuclear Fuel and Waste Management Company (SKB).

In Finland, nuclear power was far less politically controversial than in Sweden. The waste management issues could, therefore, be addressed to a larger extent from economical and practical considerations. The Finnish system has been a once-through cycle using fresh uranium. Apart from a research reactor, there has never been any other objective with building reactors than electricity generation. There have never been any large-scale uranium mines, no fuel fabrication and no reprocessing facilities.

For the first reactors in Loviisa, the management of the spent fuel was not an issue. The Russian supplier and the reactor owner Imatran Voima Oy, IVO (now FPH) made a long-term fuel management contract, which included sending back of the spent fuel assemblies to the Soviet Union and later on to the Russian Federation. This continued until the Finnish Parliament in an amendment to the Finnish Nuclear Energy Act forbade all export and import of nuclear waste in 1994 (Anttila, 2000). The last fuel shipment from the Loviisa power plant was carried out in 1996. The fuel assemblies are now stored on site. Apart from the reactors' reloading pools, there are two storage facilities in close connection to the

reactors, one with two pools and the other one with three pools. The storage capacity is 3,000 fuel bundles equivalent to 375 tonnes of uranium. Fitting the present pools with high-density fuel racks is presently expanding the storage capacity.

Teolisuuden Voima Oy, TVO, originally considered reprocessing as an option and was negotiating a contract during the construction of the Olkiluoto reactors. However, they considered the costs to be too high. The fuel is currently stored on site in a special facility, TVO KPA-store, awaiting final disposal. At present, KPA-store has three pools plus a spare pool, but is under expansion. The current storage capacity is 1,200 tonnes of uranium (Posiva, 2008).

The research reactor spent fuel that is stored in Norway comes from four heavy water reactors. JEEP I and NORA operated between 1951 and 1967, and 1961 to 1968, respectively. JEEP II started in 1966 and is presently in operation. The Halden boiling heavy water (HBWR) reactor opened in 1959 and is still in operation. At early stages in the Norwegian nuclear programme, reprocessing was still considered an option. Part of the fuel from JEEP I was reprocessed in a pilot plant at Kjeller. The first core from Halden was stored after irradiation, while the second core was reprocessed in Belgium in 1969. Since then, all fuel has been stored in Norway. The total amount of spent nuclear fuel is 16 tonnes, of which 6 tonnes is stored in IFE and 10 tonnes in Halden. Of this quantity, there are 10 tonnes of metallic fuel. This fuel is the oldest fuel and does not require active cooling. At the Halden reactor, there is a storage pool and a dry storage facility. The old metallic fuel is the dry storage. At IFE, the JEEP I and NORA spent fuel is dry-stored in vertical pits. The fuel from JEEP II is first pool-stored and cooled and then transferred to dry storage in vertical pits (Oberländer *et al*., 2009; Statens strålevern, 2003).

13.3.1 Disposal of operational waste and waste from research reactors

Finland, Norway and Sweden have repositories in operation for LLW and ILW. In 1983 the Swedish nuclear power companies received permission from the government to construct and operate a repository for short-lived LLW and ILW. This repository was built near the reactor site in Forsmark. It has been in operation since 1988 and was at the time the first of its kind in the world.

The repository, which is called SFR, is located at a depth of 50 m under the bottom of the Baltic Sea (Fig. 13.2). It consists of four rock vaults and a 50 m high, 25 m diameter concrete silo. The space between the silo and the host rock is filled with bentonite clay to avoid water flow outside the silo. It is used for the ILW, while the vaults are used for the LLW. The current

capacity of the facility is 63,000 m^3 and it receives about 600 m^3 a year. It is at present filled to half its capacity. SFR also receives RAW from hospitals, industry and research laboratories. This waste, which is first conditioned at Studsvik, constitutes only a small fraction of the total waste, typically 10–20 m^3 per year. The main types of waste containers are 200 litre steel drums, concrete or steel moulds (cubes with 1.2 m sides) and concrete tanks ($3.3 \times 1.3 \times 2.3$ m). The matrix for solidification is either concrete or bitumen (Riggare and Johansson, 2001).

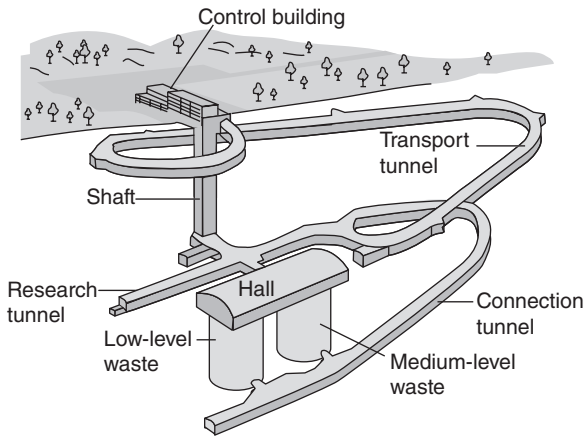
Today, there is a need for an additional capacity of 20,000 m^3 to accommodate the decommissioning wastes from the dismantling of the reactors at Barsebäck, Ågesta and Studsvik (Fig. 13.1). An additional 100,000 m^3 will be necessary for the decommissioning waste from the remaining Swedish reactors. This, however, will not be needed until some time between 2030 and 2045. The current plans are to submit an application for the extension of SFR by 2013 and to have the extended repository in full operation by 2020.

In Finland, each power company takes care of its own reactor waste. Teolisuuden Voima's (TVO) repository in Olkiluoto consists of two vertical silos at a depth between 60 and 100 m below the surface (Fig. 13.3) (Äikäs and Anttila, 2008). One of the silos is for ILW and the other for LLW. The repository has the capacity to accommodate the volumes of waste produced during the 40 years of expected operation of the existing plants at Olkiluoto. The silo for ILW has a concrete lining. Both silos are 34 m high with a diameter of 24 m. The annual amount of reactor waste is in the order of

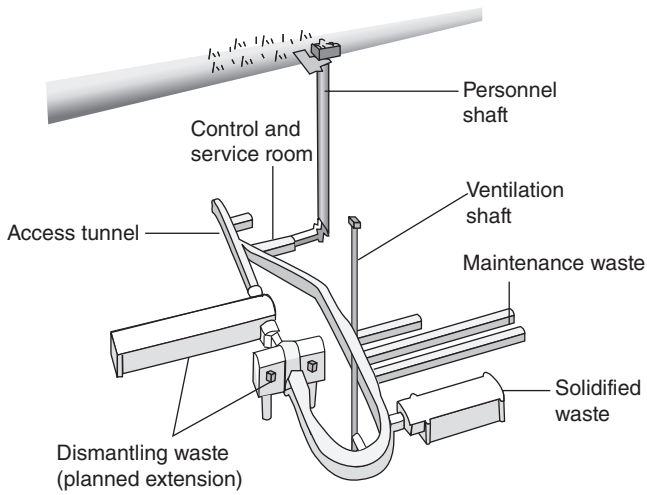
13.2 Photomontage showing the Forsmark reactor site and the underground parts of the SFR repository in Sweden. Existing parts to the right and the planned expansion to the left (from SKB's Brochure 'SKB bygger ut SFR'. Illustrator: LAJ Illustration, Photographer: Lasse Modin).

100–200 m^3. Compressible waste is packed into 200 litre drums using a hydraulic press. The drums are then compacted into half the original length. Non-compressible waste is packed into steel or concrete boxes or waste drums. The intermediate-level ion-exchange resins are solidified in bitumen and packed into drums (Äikäs and Anttila, 2008; Posiva, 2010).

The Loviisa repository has two tunnels 106 m long with a cross section of 30 m^2 for solid LLW. The ILW is deposited in an 84 m long cavern with a cross section of 300 m^2 (Fig. 13.4). The repository is at a depth of 110 m



13.3 The layout of the reactor waste repository at Olkiluoto in Finland (from www.posiva.fi).



13.4 The layout of the reactor waste repository at Hästholmen, Loviisa in Finland (from Äikäs and Anttila, 2008).

below the surface. Dry maintenance waste is packed in 200-litre drums and if the waste is compressible it is also compacted. Spent ion-exchange resins and bottoms sludges from evaporators are stored in tanks in liquid waste storage. A facility for solidifying this waste in cement was completed in 2007 (Äikäs and Anttila, 2008; Posiva, 2010).

In 1989 the Norwegian government instructed a committee to investigate the possibilities for disposal of LLW and ILW in Norway. This resulted in 1992 in an impact assessment for three possible sites with the outcome that a site at Himdalen was recommended. Himdalen is situated 25 km from the waste treatment facility at the Institute for Energy Technology (IFE) in Kjeller. It was later decided that the facility should serve for final disposal of LLW and ILW and as storage during its operational time for some plutonium-bearing waste. At the time for closure of the repository, it will be decided if that waste should be removed or conditioned in concrete and disposed of. The operation of the facility started in 1999. The repository consists of four rock caverns accessed by a 150 m tunnel. The rock cover is 50 m. The waste is packed in 210 litre drums. The total capacity of the facility is 10,000 drums. The drums are put in a concrete 'sarcophagus' and encased in cement. High-level waste and nuclear fuel will not be disposed of in Himdalen (Sörlie, 2001).

By 1970 waste barrels were being disposed of at IFE in a 4 m deep trench covered by a 2 m thick clay layer. Apart from this clay layer, this disposal facility has no engineered barriers. The current plans are to retrieve this waste, condition it and dispose of it in the Himdalen facility (Sörlie, 2001).

13.3.2 Site selection for repositories for spent fuel

The siting of a Swedish repository for spent nuclear fuel was a process that took nearly 20 years (SKB, 2011). As for the fuel management, the Stipulation Act of 1977 started the activities. The law required that the reactor owners show how and where the nuclear waste could be safely disposed of before reactors could be fuelled. From 1977 to 1985, SKB and the National Council for Radioactive Waste (PRAV, which existed between 1975 and 1981) performed site studies at eight locations, referred to as 'study sites'. Other sites were also investigated, but in some cases the local resistance was so great the studies had to be interrupted (see Fig. 13.5). Nevertheless, the investigation generated a large body of data. The main result of the study site investigations was that it is possible to find many places in Sweden where the geological conditions are suitable for building a repository for spent nuclear fuel. An important conclusion from these studies was that suitability of a site is related mainly to the local conditions and not so much to a special geological environment within the bedrock. Another lesson was that the acceptance of the local population is essential for the siting work.

13.5 Places in Sweden where investigations were conducted into potential repository sites during the period from the mid-1970s until 1990 (from SKB Report R-11-07).

About 1990, SKB established the Äspö Hard Rock Laboratory (Äspö HRL) near the Oskarshamn nuclear reactors. Collecting more data from study sites was considered not to add much to the existing knowledge. Instead it was important to develop, demonstrate and test the KBS concept and the investigation methods to be used in evaluating sites for a possible repository for spent nuclear fuel.

The stage in the siting process, referred to as feasibility studies, started in 1992 and was completed in 2000. During that period SKB approached more than 20 municipalities to discuss the possibilities of performing feasibility studies there (see Plate IV between page 448 and 449). In eight of those, SKB performed feasibility studies. The first sites to be studied were Storuman and Malå. The studies showed that both sites had favourable

conditions, but after local referenda these counties decided not to participate in further studies. In parallel with these studies, SKB also explored the possibility of siting a repository in municipalities that already had nuclear installations. In Kävlinge (Barsebäck NPP), the geology was deemed unfavourable and in Varberg (Ringhals NPP), the municipality declined to participate. Studies were, however, carried out in Östhammar (Forsmark NPP), Oskarshamn (Oskarshamn NPP) and Nyköping (Studsvik nuclear laboratories). Some neighbouring municipalities showed interest and studies were also performed in Tierp, Älvkarleby and Hultsfred. With the exception of Älvkarleby, SKB concluded that all locations provided favourable conditions for siting a repository. In 2000 SKB concluded that enough data were now available to enter into the next stage of site investigations. To select sites for the site investigations, the main factors considered were the quality of the bedrock, the possibilities for an industrial establishment in the area and political and popular support in that municipality.

Based on these considerations, SKB decided to include in the programme for site investigation phase the following:

- site investigation in Forsmark (Östhammar)
- site investigation in Simpevarp (Oskarshamn, this site also included the area later referred to as Laxemar)
- site investigation in the northern part of Tierp Municipality
- further study of the siting prospects in Fjällveden (Nyköping Municipality). The municipality, however, declined to participate further in the siting process.

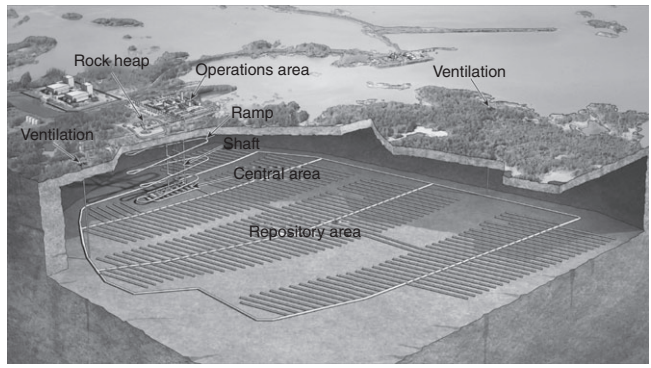
In November 2001, the government approved SKB's programme for site investigation with the exception of Nyköping, since it had already withdrawn from the process. The next step was to achieve approval to continue from the remaining three municipalities. Both Östhammar and Oskarshamn gave their approval, but Tierp declined further participation in the siting process. Following this decision SKB started site investigations in Forsmark and Simpevarp/Laxemar in the spring of 2002. Soon into the site investigation, Laxemar became the preferred site in the Oskarhamn alternative. The investigation finished in 2007 and was followed by a two-year long evaluation period.

In June 2009, SKB chose the Forsmark site as the site for a repository for spent nuclear fuel. The decisive factor was that from a long-term safety perspective Forsmark was undoubtedly the best alternative. The rock at Forsmark has fewer water-conducting fractures and also had lower permeability than the rock at Laxemar. The rock at Forsmark also provided better conditions for constructing a repository. The industrial prospects, however, for establishing and operating the final repository were good at both sites and local support for establishing a repository in the municipality was also

strong at both sites. The repository was to be designed for disposal of about 6,000 canisters. The canisters have a 5 cm thick outer copper shell for corrosion protection and a cast iron insert for mechanical stability (Fig. 13.6). These canisters will be placed in vertical deposition holes at the bottom of horizontal access tunnels and surrounded by compacted bentonite clay. A possible layout for the repository is shown in Fig. 13.7.

Following this decision, SKB submitted an application on 16 March 2011 to the Swedish Radiation Safety Authority (SSM) and to the Environmental Court in Stockholm to build a nuclear fuel repository in Forsmark and an encapsulation plant in Oskarshamn (where spent nuclear fuel will be placed in copper canisters). The main task of the Swedish Radiation Safety Authority is to decide whether the facilities comply with the Swedish

13.6 SKB's reference canister. The canister is about 5 m long with a diameter of about 1 m. The maximum weight is about 27 tonnes with full fuel load (from SKB Report TR-10-14. Illustrator: LAJ Illustration).



13.7 Repository for spent nuclear fuel in Forsmark, fully built-out (from SKB Report R-11-07).

Nuclear Activities Act with regard to nuclear safety and radiation protection. The Environmental Court will judge the applications on the basis of Sweden's Environmental Code.

In Finland, the site selection process started more than 30 years ago. After direct disposal of spent fuel had been selected by TVO as the option for management of high-level waste, systematic studies of the feasibility of geological disposal were started in 1978 and the first safety analysis for a spent fuel repository was published in 1982 (Anttila *et al.*, 1982). At that time, all the work towards a repository was performed by TVO since, according to the original agreement, the Loviisa NPP was to return all spent fuel to Chelyabinsk in Russia.

In 1983, the Finnish Government made a decision in principle that the final solution in Finland will be deep geological disposal. This meant a start in 1983 on a more focused research and development programme at TVO for deep geological disposal, including site selection. The programme aimed at selecting a site in 2000 and comprised three stages (McEwan and Anttila, 2000):

- site identification surveys (1983–1985)
- preliminary site investigations (1985–1992)
- detailed site investigations (1993–2000).

The investigation surveys aimed at identifying suitable sites for preliminary site investigations. These sites were sites with suitable bedrock for constructing a geological repository. From that pool, a small number of sites deemed to be the best suited were selected for detailed investigations.

The initial site identification used geological factors for identifying suitable areas. The selection was based on satellite photos, geological and geophysical maps. This led to a selection of 327 regions. Having defined these

regions, environmental and transport factors were then applied to reduce their number and areal extent. These factors were of two kinds, exclusionary and desirable (but not mandatory). The exclusionary factors concerned groundwater controlled areas, areas with high population density and national environmentally protected areas. The desirable factors concerned mainly transportation issues. Finland is in large parts sparsely populated with limited road and railway network in these areas, neither of which are necessarily designed for heavy transports. Another factor to be considered was land ownership. Areas with one or few owners were considered preferable to areas having many landowners.

After this selection process, 101 areas remained on the list. When the Ministry of the Environment examined the list, however, it reduced the number of investigation areas to 84 based on yet unpublished regional plans, and to that number was added Olkiluoto (the site of the Olkiluoto NPP). The Finnish Radiation and Nuclear Safety Authority (STUK) also reviewed the site selection programme and concluded that the investigation areas selected for preliminary site investigations should represent as much as possible the different geological environments in Finland.

During the review process, TVO had already started discussions with many municipalities. The starting-point was to rule out municipalities where strong opposition could be expected. Another important issue was associated with the rights of the landowners and TVO decided to consider, if possible, only areas with one landowner. In early 1986, TVO announced that an area in Ikaalinen Municipality was to be the first site for preliminary site investigation. The plan was to gradually increase the number of sites to between five and seven. Although the Municipal Counsel had encouraged the site study, opposition grew rapidly and after only a few weeks TVO decided to pull out of the area.

The experience from Ikaalinen led TVO to more actively communicate with the public in the municipalities. Issues like long-term safety and possible benefits to the community were highlighted. Following this approach, in 1987 TVO could announce a list of five sites that also satisfied STUK's request for geologic versatility. These sites were (see Fig. 13.8):

- Romuvaara (Kuhmo Municipality)
- Veitsivaara (Hyrynsalmi Municipality)
- Kivetty (Konginkangas Municipality)
- Syyry (Sievi Municipality)
- Olkiluoto (Eurajoki Municipality).

In 1992, TVO published a summary of the results from the site investigations. After this phase in the siting process, Veitsivaara and Syyry were discarded, since they were considered less suitable than the remaining three sites.

13.8 Locations of the site investigations in Finland (from SKB Report P-10-46).

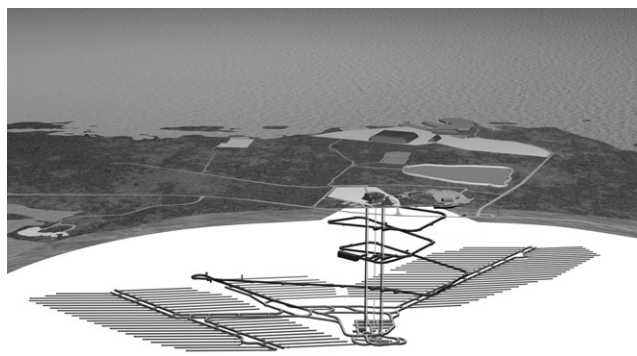
TVO intended to study the remaining three sites in more detail during the period 1993 to 2000. The overall goal was to be able to select at least one site for a spent fuel repository by 2000. After the new Finnish Nuclear Energy Act, Loviisa NPP could not ship its spent fuel to Russia. Spent fuel disposal in Finland was no longer a task for TVO alone. A site for disposal of spent fuel from FPH's Loviisa reactors had to be found in Finland. Therefore, in 1995 TVO and FPH formed a joint company for nuclear waste management, Posiva Oy, which from that time was responsible for the siting, construction and operation of a spent fuel repository. A large amount of geological data was already available at the FPH's reactor site on

Hästholmen and a pre-study showed a favourable result. Hästholmen was, therefore, included as a fourth candidate site in investigation covering the period 1997 to 1999.

At the end of the site selection programme in 1999, Posiva submitted an application according to the Nuclear Energy Act for the Government's Decision in Principle (DiP). In this application, Posiva stated that they had decided to proceed with further studies only at Olkiluoto. After a hearing process led by the Ministry of Trade and Industry, the government was able to make the DiP in December 2000, which was ratified by the parliament in May 2001. The DiP process included a legally binding acceptance by the Eurajoki Municipality to host the repository. It should be noted that this legal decision to host the repository cannot be changed in future. In 2003, Eurajoki Municipality gave Posiva a building permit for the underground characterization facility ONKALO. The work, which is still ongoing, started in 2004. Posiva's current plan is to submit an application for a construction licence for turning ONKALO into the final disposal facility (Fig. 13.9) in 2012 (Posiva, 2010). The waste packages to be disposed of are essentially the same as in the Swedish repository, i.e., canisters with a 5 cm thick outer copper shell for corrosion protection and a cast iron insert for mechanical stability (Fig. 13.6). The length of the canister differs between the two countries due to different fuel types at the nuclear power plants.

13.4 Contaminated site clean-up experience

The Swedish effort to become self-sufficient in plutonium and uranium production left installations behind that needed to be decommissioned and



13.9 Layout of spent nuclear fuel final disposal facility at Olkiluoto (from www.posiva.fi).

remediated. In 1988, the Swedish Parliament made a law regulating the financing and the responsibilities for cleaning up after the activities by AB Atomenergi, later Studsvik AB. Following that law, AB SVAFO was founded. SVAFO is currently in charge of all decommissioning work with the exception of sites owned and used by commercial nuclear power plants. These sites include the Active Central Laboratory (ACL), Ranstad uranium mine and the Ågesta nuclear reactor.

ACL was originally intended for research and development of reprocessing and production of MOX fuel, although over the years it also hosted other nuclear activities. The laboratory was opened in 1963 and closed in 1997 (Johnsson *et al*., 2004). SVAFO acquired ACL and the ventilation and filtering building (AFC) in 1998. SVAFO decided to go for complete decommissioning with the ultimate goal of demolishing the building. The laboratory was contaminated mainly with Co-60, Sr-90, Cs-137, H-3 and transuranium elements. The work started in 1998 and was completed in 2005 when SVAFO sent an application for 'free-release' of the building to the Radiation Protection Agency (SSI, now Swedish Radiation Safety Authority, SSM). The buildings were finally demolished in 2006 (Hedvall *et al*., 2006; Johnsson *et al*., 2004).

The Ranstad uranium mining and processing facility was built between 1960 and 1965. It was test operated between 1964 and 1969. The geology is alum shale with uranium content of about 300 g per tonne. During the operations, 215 tonnes of uranium was obtained from 1.5 million tonnes of alum shale. With falling uranium prices during the 1960s, the mining was not profitable and after Sweden signed the non-proliferation treaty in 1968, there was no longer a need for a domestic production of uranium. The mine was an open pit mine 2 km long, 100 m wide and 10–15 m deep. The uranium had been extracted using sulphuric acid with a gain of up to about 60–70%. Consequently, there was a large amount of uranium left in the mill tailings, which amounted to 1,000,000 m^3 covering an area of 230,000 m^2 (Stridlund and Aquilonius, 1999a).

When the mining permit expired in 1984, planning for the remediation started and was carried out during the period 1990 to 1992. The mill tailing deposit had natural stable slopes and had previously been covered by a thin layer of moraine. This layer was now covered with a sealing layer of clay-moraine mixture. A layer of crushed limestone above the sealing layer created a drainage layer. On top of this is a protective layer of 1.5 m moraine. The overall purpose was to prevent oxygen and water from reaching the mill tailings and thereby stop the leaching of metal into the water system in the environment.

When the mill tailings were placed in the area, two lakes were formed. The first lake was designed to collect the leachate from the depositions. The water was then transferred to a purification plant where it was treated with

lime to precipitate leached metals, which were deposited in a sedimentation pond before discharging the water to the second lake. That lake is now called Blackesjön. Following the restoration, the water quality was monitored in a number of locations to verify the function of the remediation system. Lake Blackesjön is now connected to the existing natural water system since the set environmental goals have been reached (WSP Environmental, 2005).

The pumping of the open pit mine ceased in 1990 and it has now been transformed into a lake, Tranebärssjön. The lake bottom is backfilled limestone and alum shale, covered by a thinner layer of backfilled moraine (Stridlund and Aquilonius, 1999b). The shoreline has been smoothened and there is a natural growth of vegetation in and around the lake. The lake and the adjacent wetland has become a sanctuary for a large number of bird species.

13.5 Problematic cases and lessons learned

There were many lessons learned from the siting process in both Finland and Sweden. During the early years following the start of the programmes, almost all emphasis was on technical and scientific issues. This was understandable since the first goal was to develop a method and a system for nuclear waste management that could be accepted as a safe solution by the scientific community. When the feasibility studies started, in Sweden in 1992 with the study in Storuman and in Finland already in 1986 with the preliminary study in Ikaalinen, it became obvious that a new dimension had been added, communicating technical issues and assessment of risk to the general public.

In Storuman, the publicity was from the beginning characterized by polarization. National actors, such as Greenpeace, came to help the local opposition and could deliver clear and well media-adjusted messages. It soon became clear that communicating the risks of a nuclear waste repository required much work and time. Communication risk using information campaigns can be a successful method if the risks are known and to some extent accepted. In the case of nuclear waste, where risks are debated, there is room for interpretation. SKB soon realized that it was essential to gain the confidence of the majority of the public in a municipality, i.e., to be seen as honest and reliable before SKB's risk assessments could be accepted. A similar lesson was learnt by TVO in Ikaalinen.

When the feasibility studies were completed, SKB's CEO summarized the lessons learned and the way forward as (Nygårds *et al.*, 2003):

- The process itself must be well known and clear to get acceptance.
- The actors/stakeholders must also see the possibilities for how or in what way the process can be affected or changed and what is fixed.

- Openness and clarity in statements from all actors is absolutely essential.
- All actors in the process must be prepared to answer questions.
- All actors must be prepared to listen to (and learn by) the arguments brought up during the process.
- Discussion in small groups and with the people potentially most affected is the most valuable part of the process to build trust and to learn about key questions.
- There will never be consensus regarding all questions. The fact that you have a consultation process does not mean that consensus will be or will have to be reached.
- The attitudes among those working in the process must reflect their belief that dialogue and discussion of these questions will create a better repository – both technically and socially.
- There must be respect for all stakeholders and their arguments and a willingness to listen and learn.

In Finland, the legally binding DiP included technical concept and public acceptance of the spent fuel repository to be located in Eurajoki. In the Finnish experience, their success factors for spent fuel and waste management can be summarized as (Varjoranta and Patlemaa, 2010):

- Long-term political commitment to resolve the spent fuel and waste issue.
- National strategy and discipline.
- Well-defined liabilities and roles.
- Establishment of funding system at early stage.
- Veto-right for the local community regarding hosting the repository in a stepwise licensing process.
- Regulator's strategic planning to allow development of regulatory approach parallel with R&D and in analogy with nuclear plant safety regulations.
- Well-structured, stepwise, open and defendable implementation programme using graded approach and 'rolling documents' strategy.
- Good safety culture and importance of dialogue between the regulator and the implementer based on comparable levels of technical competence.
- Transparency and engagement of public and domestic and international scientific and technical communities.

13.6 Future trends

Once a high level waste repository is in operation, the remaining step in the life of a nuclear power plant is the decommissioning and demolition.

This may not be a major challenge, as nuclear power plants have been demolished before (e.g., Yankee Rowe and Big Rock Point in USA). Both the Swedish and Finnish authorities (SSM and STUK, respectively) require that the reactor owner submit plans for decommissioning of their reactors. In Finland, plans were submitted in 2008 (TVO, 2008; Kallonen *et al*., 2008) and an update is due in 2014. In Sweden this is done every third year when submitting the research, development and demonstration (RD&D) plan (SKB, 2010). Sweden has already shut down three reactors, Ågesta and Barsebäck 1 and 2. The decommissioning work is scheduled to begin around 2020. In Finland, where all reactors are still operational, the stating time is about ten years later.

13.7 Acknowledgement

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13.8 References

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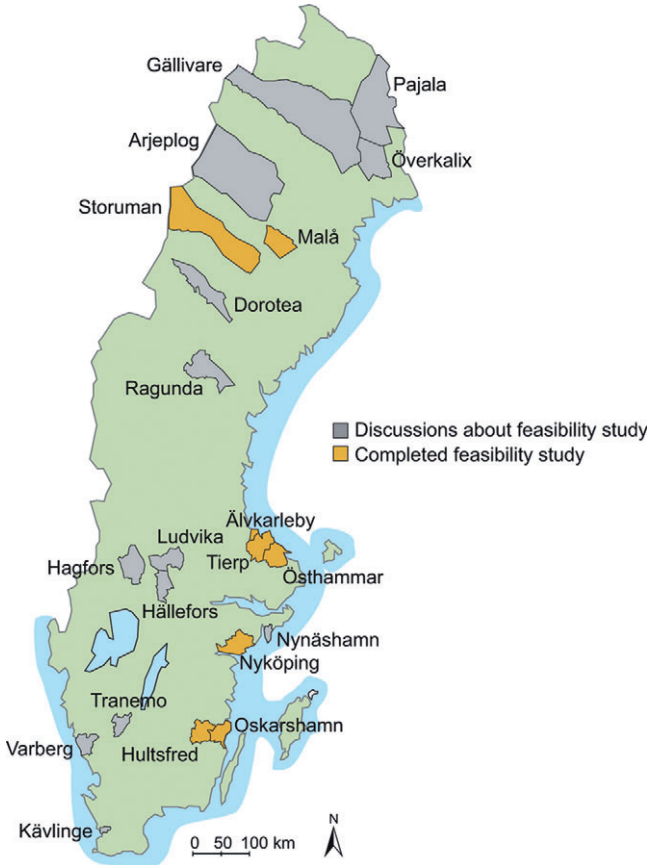

Plate IV (Chapter 13) Counties where SKB considered feasibility studies (from SKB Report R-11-07).

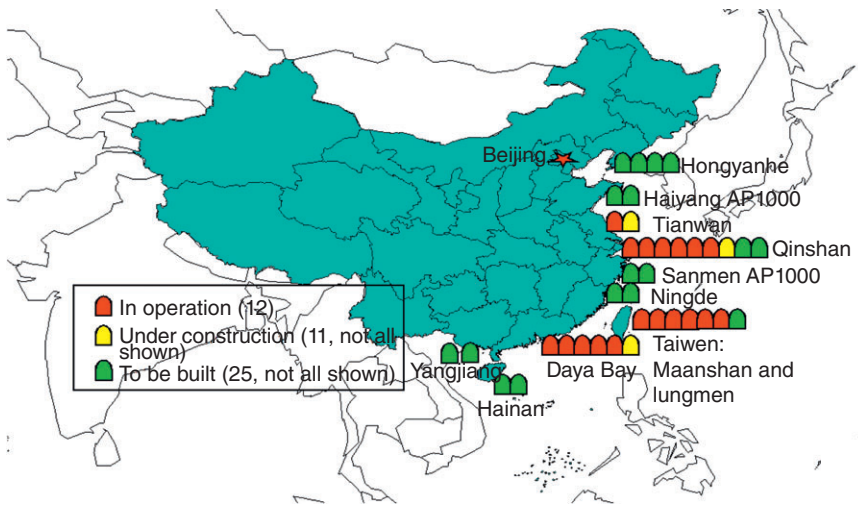

Plate V (Chapter 22) Distribution of NPPs in China.

2

Radioactive waste (RAW) categories, characterization and processing route selection

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Abstract: The principal approach to radioactive waste management is to transform 'as generated' waste to a waste package suitable for safe long-term storage or ultimate disposal. A waste characterization system allows an assessment of the potential risks connected with waste handling and disposal and also allows the waste to be classified into groups (streams) according to their properties and projected processing routes. A properly selected waste classification system also enables the selection of the proper processing technology for each class of waste, tailored to waste volume, properties and available technologies in each country or waste processing organization. Long-term safe disposal of processed waste is a basic requirement of all waste classification and waste processing schemes discussed in this chapter.

Key words: radioactive waste, properties, characterization, record keeping, waste processing routes.

2.1 Introduction

The use of nuclear energy for power generation and application of radionuclides in medicine, research, education and in several industrial fields is still increasing. Generation of radioactive waste (RAW) is an unavoidable by-product of generating nuclear energy and safe and sound waste management to ultimate disposal in environmentally safe conditions is not only a technical and technological problem, but also a serious socio-political problem, with potential direct impact on the future of nuclear energy.

Radioactive waste is generated in a wide variety of compositions, radionuclide content, physicochemical parameters, and volume. Each category of RAW requires individual management and an individual technological approach. However, there is a basic tenet for all waste processing which is to transform each waste into a waste package suitable for safe isolation either in storage or in a final disposal facility to ensure that any impact on the population and environment is as low as possible.

Many classification systems have been developed and implemented in the more than 60 years of the nuclear era. Most of them are based on the radiological characteristics of the waste. However, the classification systems are usually tailored to adhere to technological requirements, country-specific legislation, as well as national waste management policy requirements and limitations. The International Atomic Energy Agency (IAEA), as world co-ordinator of nuclear issues, facilitates dissemination and exchange of information, 'know-how' and good operational practice in waste classification. This makes a big difference in the harmonization of approaches at an international level; in particular, in the proposed general RAW classification scheme, depending primarily on the final disposal options being considered in relation to the radiological parameters of the waste. This scheme can be tailored according to the individual requirements of the various countries or even the various waste management organizations. However, even though the classification scheme is in a generalized form, it allows the intercomparison of various classification systems and is often used to facilitate the transfer of RAW management 'know-how' among the international community.

Based on a generated waste volume and the characteristics of a waste, a proper and safe waste processing technology can be selected. At present, there are safe and proven technologies available for processing of all kinds of RAW. The main issue is, therefore, to select an optimum technology or a sequence of technological procedures to produce a waste package that corresponds to the safety requirements for long-term storage and ultimate disposal, while at the same time considering the operational staff radiation protection, economic issues, public perception and other aspects.

All these issues are discussed in this chapter as a general introduction to the more technologically oriented chapters in this book.

2.2 Basic categories of radioactive waste (RAW)

Radioactive waste (RAW) is generated in various activities and in a number of different facilities. The most significant source of RAW is nuclear energy generation; in particular, operation of nuclear power plants (NPP) and the disposition of the spent nuclear reactor fuel, i.e. whether the fuel is recycled or considered as waste. Other sources are so-called institutional applications of radioisotopes: medical, research, educational, industrial and other facilities. The origin of the RAW usually also determines its basic characteristics and represents the principal information necessary for its categorization and classification as well as the information necessary for consideration and decision making on how the RAW is processed and the potential disposal routes.

There are several ways to characterize and categorize RAW. Besides chemical composition, aggregate state, mechanical properties, etc., the main characteristics are radiological properties, namely the activity concentration and the type and physical parameters of the radionuclides in the waste.

This book is aimed mainly at NPP operational and decommissioning radioactive wastes and spent fuel (if considered as a waste) management, with emphasis on technological aspects of RAW management. The aim of this chapter is to describe the categorization of these wastes.

2.2.1 Spent nuclear fuel (SNF)

Spent nuclear fuel (SNF) contains the major portion of the radioactive material generated in NPP. The SNF contains most of the highly radioactive fission products generated in a reactor as well as significant amounts of transuranium elements (TRU), generated in neutron activation of non-fissionable bulk fuel material and low activity bulk fuel material (e.g., uranium oxide). Under normal conditions fuel element cladding material should provide a tight barrier, providing for sufficient separation of the radioactive material from the environment during the entire fuel post-irradiation lifetime.

The SNF is the only nuclear fuel cycle material that can be considered either as radioactive waste or as a valuable source of fissile material. The decision between these options is usually not sharp and is based on various countries specific technical, technological, political, strategic and other considerations. It is common that the same type of SNF in one country may be slated for reprocessing and in another country it may be slated for final disposal. The following two options of SNF categorization also can influence which management approach is applied:

- SNF is a valuable source of secondary fissile material (if a closed nuclear fuel cycle is applied). Secondary fissile material (in particular, Pu-239) together with non-used fissionable U-235 is separated from SNF in reprocessing facilities and used in the manufacturing of fuel for particular types of nuclear reactors.
- SNF is considered and managed as RAW (if an open nuclear fuel cycle is applied). After proper cooling and packaging, SNF should be disposed of in deep geological repositories. If a deferred decision on disposal is considered or if a disposal facility is not yet available, then long-term storage under strictly controlled conditions must be applied.

SNF, as a potential source of nuclear weapon material, is fully covered by the IAEA safeguard rules and guarantees, which have to be obeyed and respected in each SNF handling and management step.

2.2.2 NPP operational and decommissioning RAW and institutional RAW

The NPP operational radioactive waste category incorporates the waste generated in both the operation and decommissioning of NPPs. RAW generated in institutional applications are also included in this group and in some countries are even managed (entirely or only for some steps) together with NPP waste. This category is the biggest from the point of view of the waste volume generated and covers a wide spectrum of wastes with respect to their radiological, chemical, physical, aggregate and other properties. Therefore the classification system is complicated, usually reflecting country-specific approaches, national waste management strategy, regulations, and often technological and other aspects. The IAEA in its Radioactive Waste Management Status and Trends [1] has recognized more than 40 different country-specific RAW classification systems. It is obvious that under such conditions it is difficult to unify approaches, evaluate and compare national systems, and make a plan for optimization and recommendations for safe RAW management.

The IAEA has made a systematic effort to develop a unified and internationally accepted classification (categorization) of RAW for a long time in order to facilitate communication between involved parties in various countries and organizations. At the beginning of the 1990s the first comprehensive IAEA document was published on Classification of Radioactive Waste [2], which based the RAW classification system on aspects related to waste disposal safety and proposed the boundaries of individual RAW categories using International Commission on Radiological Protection (ICRP) recommendations for annual individual and collective doses for public. The two main radiological parameters used for the RAW classification system are the radioactive material concentration and the half-life of critical radionuclides. This proposed system has been accepted for development of a national RAW classification system in many countries.

Further evolution resulted in a new IAEA RAW classification system published in 2009 [3], which is more generic and almost exclusively based on long-term waste disposal safety considerations (in other words, for each RAW class there is a specific proposed/assigned waste disposal route). This recent waste classification system was modified in order to reflect a better relationship between RAW categories and the safety aspects of the considered disposal options. While the 1994 IAEA document provided basic numerical boundary values for various RAW classes, the 2009 document offers only a general approach and leaves the development of more exact figures to the individual national regulations. For illustration and a better understanding of the boundary parameters and values for distinction of the

RAW categories, the data from the 1994 IAEA document [2] are given in Table 2.1. In spite of the fact that the 1994 IAEA document [2] is no longer applicable, while the updated IAEA 2009 document is available [3], these figures are still used as informative values in preparation of the national classification systems.

In accordance with the present IAEA approach to RAW categorization [3], six classes of waste are proposed for consideration in preparation of the national classification systems. It should be noted that precise values of applicable total activity content and eventually activities (activity concentrations) of the most significant individual radionuclides for each waste category shall be specified on the basis of safety assessments for individual

Table 2.1 Typical characteristics of waste classes [2]

Waste classes	Typical characteristics	Disposal options
1. Exempt waste (EW)	Activity levels at or below clearance levels given in IAEA draft document[a], which are based on an annual dose to members of the public of less than 0.01 mSv	No radiological restrictions
2. Low and intermediate level waste (LILW)	Activity levels above clearance levels given in IAEA draft document[a] and thermal power below about 2 kW/m^3	
2.1. Short-lived waste (LILW-SL)	Restricted long-lived radionuclide concentrations (limitation of long-lived alpha emitting geological radionuclides to 4000 Bq/g in individual waste disposal facility packages and to an overall average of 400 Bq/g per waste package)	Near surface or geological disposal facility
2.2. Long-lived waste (LILW-LL)	Long-lived radionuclide concentrations exceeding limitations for short-lived waste disposal facility	Geological disposal facility
3. High level waste (HLW)	Thermal power above about 2 kW/m^3 and long-lived radionuclide concentrations exceeding disposal facility limitations for short-lived waste	Geological disposal facility

[a] IAEA draft document: IAEA Clearance levels for Radionuclides in Solid Materials: Application of Exemption Principles, Safety Series No. 111-G-1.5, in preparation in 1994, IAEA, Vienna. Later reconsidered, rewritten and published as Reference [4].

disposal route and disposal site. This is the responsibility of the national authorities and the values used can differ significantly in individual countries.

Exempt waste (EW)

Exempt wastes are the wastes that meet the criteria for clearance, exemption or exclusion from regulatory control for radiation protection purposes as described in Refs [4] and [5]. The concentration of radionuclides in exempt wastes is negligibly small and no provisions are required for radiation protection of professional staff and public, irrespective of the disposal route (RAW disposal facilities or common conventional landfills). No special requirements are established for management and disposal of EW, and for RAW managers and for technologists it is always questionable whether EW should be considered in RAW management planning, or whether such wastes could be omitted and managed as non-radioactive waste. The exemption procedure, consensual criteria for exempt waste and exemption levels for total activity and activity concentration of individual radionuclides are established in Refs [4] and [5]. They are based on dose rates for the public, recommended by the ICRP and generally accepted worldwide.

Very short lived waste (VSLW)

RAW containing radionuclides with very short half-lives can be stored while it decays over a limited period of up to a few years and is subsequently cleared from regulatory control according to arrangements approved by the regulatory body. This category typically includes 'monoisotopic' RAW coming from institutional applications of radionuclides, in particular, in medicine and research. Proper storage conditions should be arranged to ensure safety during the decay period.

Very low level waste (VLLW)

The VLLW category comprises the wastes for which the activity concentration is slightly higher than that required for EW, but VLLW does not need a high level of containment and isolation. Landfill type facilities with limited regulatory control, used, for example, for other non-radioactive hazardous waste, can be used for disposal. A typical source of such waste is decommissioning of nuclear facilities, when large volumes of very low contaminated soil, rubble, concrete, thermal insulation, etc., are generated. Concentrations of long-lived radionuclides (e.g., nuclear fuel components) in VLLW are generally very limited.

Low level waste (LLW)

This category comprises the wastes with radioactivity concentrations well above clearance levels, but with limited amounts of long-lived radionuclides. A typical source of this waste is NPP operation and 'as generated' waste in various physical forms and of various chemical and radiochemical composition. Various radionuclides (fission products, activated products) are present in a wide scale of activity concentrations. Such waste requires robust isolation during handling and storage and containment for periods of up to a few hundred years (300 years is typically considered, based on ten times the Cs-137 half-life). After proper processing and containment, it is suitable for disposal in engineered near-surface facilities.

Intermediate level waste (ILW)

Intermediate level waste is the waste that, because of its higher radioactivity concentration and/or higher concentration of long-lived radionuclides, does not fit into the previous LLW category. ILW may contain long-lived radionuclides, in particular, long-lived fission products and alpha emitting radionuclides that will not decay to a level of activity concentration acceptable for near-surface disposal during the time for which institutional controls can be relied upon. However, ILW needs no provision, or only limited provision, for heat dissipation during its handling, storage and disposal. The activity concentration of bulk radionuclides, as well as minor long-lived radionuclides used to distinguish between LLW and ILW are not universally agreed upon. Moreover, the upper activity concentration limits for ILW are not universally agreed upon. These limits and concentrations are site-specific and they shall be established in each individual case by the regulator, based on a safety analysis of the disposal option being considered. Some guidelines about the limiting values for long-lived radionuclide activity concentrations can be found in Table 2.1.

ILW is typically generated at NPP as a result of treatment (concentration) of primary waste. Another significant source is reprocessing of spent fuel. ILW requires a greater degree of containment and isolation than LLW and disposal in subsurface repositories at depths of the order of tens of metres to a few hundred metres.

High level radioactive waste (HLW)

High level waste is waste with levels of activity and radionuclide concentrations high enough to generate significant quantities of heat by radioactive decay or waste with large amounts of long-lived radionuclides that need to be considered in the selection of a disposal facility and disposal route for such waste. Handling and storage of HLW requires proper shielding and in

some cases also additional cooling. Typical examples of HLW generated at NPPs are highly activated reactor parts. However, the main source of HLW is reprocessing of spent nuclear fuel. Disposal in deep, stable geological formations usually several hundred metres or more below the surface is the generally recognized option for disposal of HLW.

2.2.3 Disused sealed sources (DSS)

DSS are a special category of institutional RAW coming from various industrial (non-destructive testing), medical, research and other applications. Under normal conditions, they comprise firmly-contained single radionuclides with an activity that can vary over several orders of magnitude – from low dose rates brachytherapy and positron emission tomography (PET) sources (typical activity of 10E-2–10E-4 TBq) to highly active teletherapy sources and radioisotope thermoelectric generators (typical activity of order of magnitude 10E-4 TBq). Various radionuclides, almost exclusively artificial, are used in sealed sources. The categorization of new sealed sources, described in detail in Ref. [6], is fully applicable also to DSS. A detailed registry of sealed sources is usually established according to national regulations and the most common procedure for management of spent sealed sources is to return them to the manufacturer, who is responsible for further disposal.

2.2.4 RAW from fuel cycle front end (uranium and thorium mining and milling)

A huge amount of low level RAW from uranium and thorium ore mining and milling can be found in many locations worldwide. The mines and mill tailings are often abandoned and RAW, generated by mining companies, is often left on the abandoned site. Tailings and tailing ponds and their closure, stabilization and isolation from underground water are the main challenges of this category of waste management. Tailings contain only naturally occurring radionuclides and besides their extremely large volume, the main problems are caused by radiochemical (content of long-lived radionuclides, alpha emitters) and chemical composition (e.g., high acidity), which varies from site to site, depending on the mining process applied at a given site. Categorization of this type of RAW and the basic approach and strategy for their management can be found in Ref. [7].

2.3 RAW characterization and control

Reliable RAW processing shall result in a waste package, corresponding to the requirements of the waste disposal site, established usually as waste acceptance criteria (WAC).

The establishment of an efficient waste control strategy and implementation of a proper waste characterization and control system is one of the key conditions to achieve the WAC. The RAW control system shall cover control of radiological, chemical and mechanical properties of RAW in each RAW stream and during the entire RAW life cycle. There are three principal steps in RAW control:

- Characterization of 'as generated' waste, providing principal information on the waste parameters for waste processing planning and selection of processing technologies. This information is also a basis for estimation (prognosis) of the final waste package parameters.
- Control of RAW processes and operation of processing technologies (both treatment and conditioning steps), including control of material inputs, e.g. waste encapsulation matrix components, if applicable.
- Characterization of the final waste package to demonstrate its compliance with the WAC.

Commonly, RAW control is performed in different types of laboratories from simple, operational laboratories for basic waste and processes control to advanced radiochemical laboratories for precise control of critical waste parameters. The principal steps in a laboratory waste control procedure are: sampling, chemical parameter determinations, radiological parameter determinations and control of mechanical properties (applicable mostly for the final waste form and/or waste package). An unavoidable condition for advanced RAW control system implementation is the availability of a well-qualified accredited control laboratory, properly equipped with the relevant instrumentation, having in possession radiochemical methodologies for sample processing (ISO, ASTM, local accredited procedures), with a highly qualified staff and implementation of an advanced quality assurance/quality control (QA/QC) system.

The systematic or random control of finalized waste packages, whether for long-term storage or final disposal, is usually performed by non-destructive gamma or neutron scanning (tomography) of entire waste packages, using special scanning facilities, facilitated with special software for evaluation and presentation of results.

A consistent part of the waste control system is record keeping, providing traceability of waste parameters for regulatory purposes as well as for potential future decision making.

A compendium, describing the establishment of a waste control strategy, the distribution of responsibilities, a quality assurance system, and providing an overview of characterization methods and procedures is given in Ref. [8].

2.3.1 Chemical parameter control

Chemical parameter control is applied mostly in liquid waste streams and at the beginning of the waste life cycle. Operational control of selected technological equipment can also be incorporated in the control plan. Controlled parameters for each waste stream shall be carefully selected and optimized. The control plan shall be established to reflect the requirements of the technological equipment operator and to assure the quality of the final product and its compliance with the requirements for waste disposal (WAC). Information on chemical composition, acidity, salinity and other chemical and physical-chemical parameters is used in RAW process technology planning, and the 'as generated' waste parameters can be adjusted to suit the process technology. Another objective is to manage different waste streams to optimize process conditions for available processing technologies with the aim of achieving the best possible utilization of disposal facility space ('filling' the container) and at the same time assuring compliance with the WAC.

A declaration of selected biological parameters might also be required by the WAC; therefore their control shall be performed on selected waste streams (in particular in waste streams containing organic material), usually in parallel with control of chemical parameters.

Waste matrix parameters and final waste form quality control (qualification tests: chemical durability, leaching properties, long-term performance in disposal site conditions, etc.) are also a significant part of a laboratory chemical control system.

2.3.2 Radiological parameter control

Waste acceptance criteria (WAC) for disposal, a principal requirement for qualification of a produced waste package, are established predominantly on the radiological parameters of waste packages. Radiological parameter control is, therefore, considered to be the main component of a RAW control system. The WAC are country-specific; however, the IAEA recommendations for the establishment of WAC are accepted as the basis worldwide. Besides surface dose rates, maximum permitted activity concentrations (or total activity per entire waste package) of several radionuclides is usually defined in a WAC. The list of considered radionuclides is different for each country's WAC for disposal. Besides common and simple measurable radionuclides (such as Cs-134, Cs-137, Sr-90, etc.), a declaration of the activity concentration of 10–40 so-called critical radionuclides for disposal (alpha emitters, biologically important radionuclides, long-lived radionuclides usually with half-life over 30 years, etc.), is also required in a WAC. These radionuclides are often difficult to measure. To facilitate declaration

of waste package compliance with a WAC, non-destructive (mainly gamma spectrometry) as well as destructive radiochemical procedures (with radiochemical processing of the samples) are routinely applied.

Radiological control is applied in the entire life cycle of RAW. However, analogous to chemical parameters, the main effort is put on the radiological control of 'as generated' (raw) waste and then on the declaration of RAW package compliance with a WAC. For radiological control of 'as generated' waste, carefully selected combinations of non-destructive instrumental methods and radiochemical analysis with separation and subsequent determination of difficult to measure radionuclides (some fission products, transuranium elements, etc.) are applied. The information obtained is widely used in waste processing planning for each waste stream and in the estimation (prognosis) of final waste package parameters. Results of radiochemical analysis of input waste are also used for determination of radionuclide vectors, necessary for application of scaling factor methods (see below).

Most often a non-destructive check of the entire waste package is used for a declaration of final waste package compliance with a WAC. Gamma scanning, gamma tomography and in some cases also neutron tomography, all in combination with advanced data processing, are commonly used by both waste package producer as well as by disposal facility operator. The above-mentioned techniques allow determination of the major gamma-emitting radionuclides and along with using neutron tomography determines the major actinides and fissile material. In general, non-destructive determination of minor radionuclides, critical for disposal, is very complicated, expensive, and in some cases even impossible. Destructive determination with sampling of the waste form and waste package material and subsequent laboratory radiochemical analysis is not only technically complicated but can cause unacceptable damage to one or more of the waste isolation barriers in the waste package. The situation is more substantial for processed liquid waste, where critical disposal radionuclides can be expected with higher probability. The way around this situation is the application of scaling factors and a nuclide vector methodology [8]. The substance of this method is simple; however, implementation is more complicated and requires special software tools. Careful and precise radiochemical analysis of homogenized waste before the start of its processing is used to establish the nuclide vectors – a mathematical relationship between the activity concentration of major or easy-to-determine radionuclides (usually strong gamma emitters) and the activity concentrations of minor (usually difficult-to-determine) radionuclides is developed. Using nuclide vectors and thorough knowledge of the waste processing procedure and waste package materials, it is possible to calculate and declare activity concentrations of minor radionuclides in a waste package using measured data on the activity of the major radionuclides, obtained by non-destructive gamma scanning

of the entire waste package. Such a procedure should be, of course, qualified and approved by the regulator and disposal facility operator.

2.3.3 Physical and mechanical parameters

Knowledge of physical parameters of solid 'as generated' RAW is important for some processing technologies, like compaction and pyrolysis. The requirement for the content and level of information should come from the facility operator and a methodology to determine the parameters shall be tailored accordingly.

Information on the physical and mechanical parameters of processed waste is substantially more important. Demonstration of key mechanical parameters of a waste form and the entire waste package is usually required by the WAC. This requirement comes from the projected long-term durability of the waste form (in particular for solidified liquid waste) and also from the design and arrangement of waste packages in the disposal facility, where placement of waste packages in several layers is commonly used. The last requirement is usually solved by use of verified and approved waste containers, providing for sufficient mechanical stability for the entire waste package. Mechanical parameters of the waste form are controlled in the waste producer facility using samples taken during waste processing – the scope of control and methodologies should be developed according to the requirements of the WAC and the expectations/requirements of the disposal site operator.

2.3.4 Record keeping

During its processing, radioactive waste is converted from an 'as generated' state to a processed waste form and placed in a container to form a final waste package for storage and disposal. A principal condition for acceptance of waste packages for disposal is full compliance with the disposal site WAC, in other words, to demonstrate that chemical, radiochemical, biological, mechanical and other parameters of the waste form are in accordance with the required values. The waste parameters can change during handling and processing, and to ensure compliance of a waste package with a WAC, a system for generating and maintaining records should be established in order to save and track all relevant information. It is worth registering not only the waste parameters but also the technological parameters of the processing facilities. A record-keeping system should define the data, which should be collected and stored at each step of the waste life cycle and for each waste stream. A reliable selection system should be implemented not only to avoid collecting too much information, but, also to

assure the long-term availability of all significant and potentially needed data. Record-keeping systems for the pre-disposal period of the waste life cycle should ideally be coordinated and interconnected with the record-keeping system for the disposal facility. However, a reasonable data reduction approach should be applied for transfer of the information. More detailed information for the identification of requirements and establishment of record-keeping systems can be found in Ref. [9].

2.4 RAW processing route selection

The final objective of waste processing is to transform 'as generated' waste to the form suitable for final disposal, providing for high safety and avoiding any significant burden to the environment and population. Several technologies have been developed and implemented to process various types of waste and waste streams. All of them are generally aimed at reducing the original waste volume and providing sufficiently stable and durable waste forms, suitable for long-term storage and ultimate disposal.

Basically two approaches can be applied for the reduction of 'as generated' waste volumes:

1. Removal (concentration) of radionuclide contamination from the waste and processing of the small volume of concentrate as higher (intermediate) level radioactive waste. After removal of radioactive material from the waste, the bulk of the original waste volume can be managed as non-radioactive (cleared from regulatory control) or very low radioactive material at common conventional landfills, or discharged to water reservoirs (sea, river). Significant reduction of liquid waste volume can be achieved in this way. However, some complications should be expected in relation to handling and further processing of the waste concentrate as intermediate level waste.
2. Reduction of volume of 'as generated' waste (e.g., by evaporation of liquid waste or thermal treatment/pyrolysis of solid waste) for further conditioning into a waste form suitable for disposal. The waste matrix in this case represents the bulk of the processed waste volume and, therefore, more space is required in the storage or disposal facility.

Selection of a waste processing route and a decision on its implementation is a complicated process, where technical, economic, safety and other aspects as well as level of industrial development, size of nuclear industry, availability and type of waste disposal options available in a country should be considered and evaluated. Typical examples of different approaches to waste storage and disposal, leading to different waste processing approaches are, on one hand, the Netherlands, where controlled long-term (100 years) storage of processed waste in special surface storage facility is implemented,

while, on the other hand, Germany, where deep geological disposal is the only considered option for all kinds of waste. This latter approach could benefit from higher flexibility in selection of waste processing technologies. And the third, classic example is the case of several European countries, operating near-surface repositories for disposal of processed low and intermediate level waste, where strict WAC requirements must be obeyed.

2.4.1 Integrated RAW management approach

The main objective of RAW processing is to avoid any potential negative impact of the processed RAW on the population and environment for a sufficiently long time, necessary for the decay of the major fraction of radionuclides contained in the waste. This target is normally achieved by:

- selection and application of proper waste processing technology, assuring production of waste packages, corresponding to WAC;
- implementation of safe and proven long-term storage or disposal of waste packages, providing for high safety and reliability.

It is clear that long-term storage and/or permanent disposal are key issues. There is a lot of flexibility in selection and implementation of waste processing technologies; however, there is almost no chance to modify a waste package once it is already made: the waste package can either be accepted for storage and/or disposal or not. And 'not' in this context always means many problems, which are sometimes very difficult to solve.

Therefore any consideration of waste handling and processing activities should start with the end product – from detailed analyses and evaluation of available disposal conditions and requirements. The waste manager, the planned waste processing steps, should always consider the 'end-point' of the waste life cycle – disposal – and propose an integrated sequence of linked steps, following waste management policy, aimed at the production of waste packages compliant with WAC. Properly designed sequences of waste processing steps should provide for a systematic step-by-step increase of safety features related to the processed waste and, at the same time, minimization of waste volume. All parties involved in the waste management shall assume responsibilities to assure that only acceptable risks are taken.

Such a logical and preferably optimized strategy, which includes a complex set of technical and administrative measures, must be used in the planning and implementation of a RAW management programme as a whole from waste generation to disposal. The strategy must be such that the interactions between the various stages are taken into account so that

decisions made at one stage do not preclude certain alternatives at a subsequent stage: this is usually called the 'integrated approach' to RAW management [10].

2.4.2 Waste package specification and waste acceptance criteria

The *waste package* as a final product of RAW processing, prepared and ready for long-term storage, or disposal, consists of two components: the waste form and the waste container. In some cases there are also additional barriers or shielding used to increase the safety features of the waste package. The waste package should be prepared in a form corresponding to the requirements for handling, transport, storage and disposal.

A *waste form* is defined as 'radioactive waste after treatment and conditioning, usually in solid form, prior to its packaging into the waste container'. A *waste container* is defined as 'the vessel into which the waste form is placed for future handling, storage and disposal'. The waste container fulfils the role of a protective barrier and shielding tool. The waste container should guarantee the tightness for the entire period of storage and/or disposal of the waste.

To ensure the long-term safety of waste disposal, WAC should be developed based on a safety assessment of the considered disposal options and should be approved by the relevant authorities. Waste acceptance requirements (criteria) are by definition quantitative or qualitative criteria for processed RAW to be accepted by an operator of a repository for disposal, or by an operator of a storage facility for storage. WAC are specified by the relevant authorities, or proposed by an operator and approved by the relevant authorities. Waste acceptance requirements might include, for example, restrictions on the activity concentration, or the total activity of particular radionuclides (or types of radionuclide) in the waste, or requirements concerning the waste form or waste package [10]. In the past, the term waste acceptance criteria was only applied and used in the context of waste disposal. Later on, the approach to specify WAC has been extended also to some other steps of the waste life cycle – in particular, for transport and storage. In general, WAC can be specified for any foreseen waste management operation and handling. WAC can prescribe and cover various waste package features and properties, such as:

- requirements for waste packages (e.g., surface dose rates, surface contamination, mass, leak tightness);
- requirements for waste forms (e.g., radionuclides content, composition and parameters of waste matrix and solidified waste, encapsulation material);

- requirements for waste container (e.g., design features, mechanical stability, thermal resistance and also some additional features – shielding, corrosion protection, etc.);
- limitations for activity – permissible activities of individual radionuclides, total activity, activity of selected critical radionuclides;
- radiological safety parameters – surface dose rates, surface contamination.

There are usually several other requirements, developed and specified based on the safety assessment of risks of the planned operations (transport, storage, disposal, etc.) with prepared waste packages. A more detailed description of this subject can be found in Chapter 3.

WAC are *site-specific*, but *not container-specific*. They are developed based on a safety assessment of the design and implementation of the waste disposal or waste storage facility and eventually waste transport tools. WAC are general criteria, usually not specified for particular waste containers and/or waste packages. Therefore they are not simply applicable in everyday technological practice.

To overcome the above limitations of WAC, the general waste acceptance requirements are usually transformed into detailed *waste package specifications* (WPS). WPS should be developed and individually implemented for each type of RAW package and should reflect specific characteristics of the waste package. WPS are therefore waste package (and also waste container) specific and they are normally substantially more detailed than WAC. They shall be a consistent part of the QA/QC system applied by the waste package producer. Application of WPS allows simple control and verification of waste packages for both the waste producer as well as the waste disposal facility operator. Compliance of waste packages with WPS is considered a guarantee of compliance of said waste package with the WAC for a particular waste lifetime step. More details and guideline for development of WPS can be found in Ref. [11].

Waste acceptance criteria for disposal can normally be developed based on the safety assessment of an available, already constructed, or intended waste disposal facility. In any case, a clear idea of the waste disposal option should be available. However, many countries are in the situation where processing of RAW is unavoidable and the decision regarding a disposal facility is still deferred. In such cases, there are two principal options on how to proceed with waste processing to avoid future complications with acceptance of waste packages at the disposal site:

- Develop and apply generic WAC, based on international experience, approaches, and analogy with similar nuclear programmes. In this manner, a sufficiently conservative approach shall be taken and it shall be demonstrated that a national waste management policy and vision

of future disposal options had been considered. These criteria can then be used for development of waste package specifications for available waste management technologies and waste packages already in use.

- Develop and apply only waste package specifications for particular waste streams and waste processing technologies, based on a detailed analysis of potential disposal options. A sufficiently conservative approach and margins in critical parameters shall be applied to avoid future problems with acceptance of waste packages for disposal.

2.4.3 Principal steps of RAW processing

There are two principal steps in RAW processing: treatment and conditioning. The main role of treatment is to change the characteristics of the waste or to reduce the volume to make waste suitable for final processing by conditioning. The main role of conditioning is to incorporate or encapsulate the waste into the waste matrix, and/or package the waste in a container, where the container functions as an efficient and safe barrier for isolation of the waste from the environment. The distinction between treatment and conditioning is sometimes not clear – depending on the national waste management policy and approach, some kinds of treated waste can be considered as suitable for disposal and in other cases can be considered as needing further processing. This decision also depends, of course, on whether an ultimate disposal option is available or expected. More safe, advanced and sophisticated disposal options can potentially lower the requirements for the conditioning procedure, for example if deep geological disposal of low and intermediate level waste is considered, packaging of the waste into special containers can be acceptable instead of solidification into an encapsulation matrix.

In some cases pre-treatment of RAW is applied to modify 'as generated' waste into the form suitable for further treatment. A typical example is adjustment of the chemical properties (e.g., adjustment of acidity, or destruction of organic compounds in the waste) of liquid waste. Segregation of solid waste is also often considered as pre-treatment procedure.

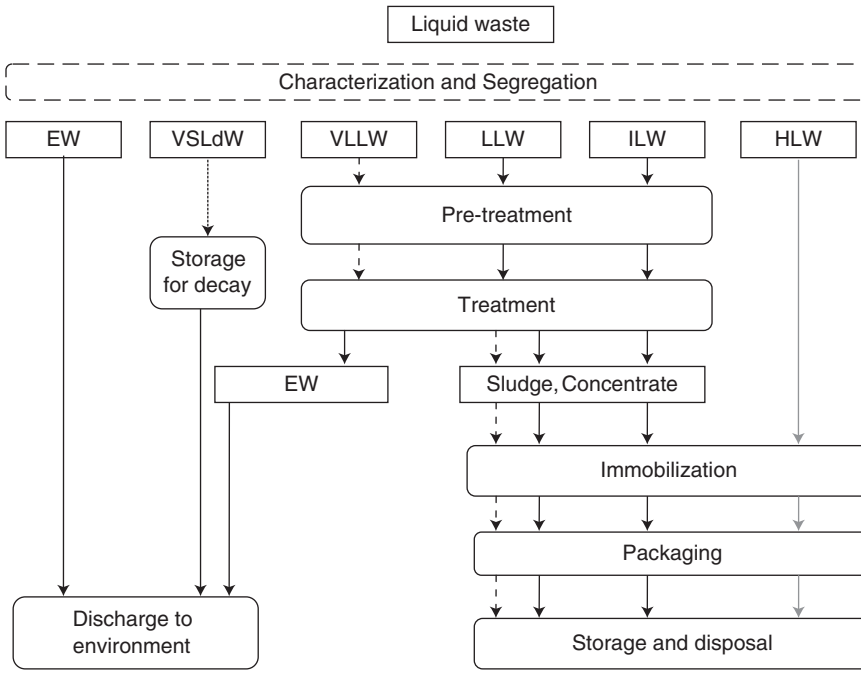
In addition to the above-mentioned classification of RAW based on its properties, technologists often use their own 'technological classification' of the waste. It is based on waste stream characteristics in combination with available technologies for their processing. Sometimes, a technological classification allows 'as generated' waste streams to be merged according to typical waste characteristics and this provides for more efficient processing. A technological classification of waste is very individual, specific almost for each type of nuclear facility. However, any technological classification should obey the basic rule – to be consonant with waste package specifications and WAC.

Considering the types, properties and volume of generated waste and taking into account the available waste processing technologies, waste processing organizations should prepare complex waste management plans to assure management that all kinds of waste and all waste streams are handled in a safe and sound manner with one goal – to produce waste packages acceptable for ultimate disposal or long-term storage, i.e. compliant with WAC for disposal or long-term storage.

The main decision-making parameters in waste processing technology selection are activity concentration and aggregate state. With regard to aggregate state, two principal categories of RAW are generated at nuclear facilities: liquid waste and solid waste. While the composition (chemical and radiochemical) and properties of liquid waste depends more on the type of reactor, the composition and properties of solid wastes do not depend substantially on the reactor type. Both primary waste streams are pre-treated and treated directly at the generator's site and the volume of treated liquid waste is usually lower than the volume of solid waste. Another, smaller volume, waste category is spent ion exchange resins and other filtration materials, sometimes admixed with radioactive sludge and/or sediments from liquid waste storage tanks. These resin, filtration materials and sludge/sediments are much more difficult to process. However, they are sometimes considered in the liquid waste category because of the high content of water and slurry. In some countries they are declared as a separate 'wet waste' category.

The principal scheme of aqueous liquid radioactive waste management is presented in Fig. 2.1, taken from Ref. [12]. Various types of aqueous liquid RAW are generated at various nuclear facilities. However, the processes by which they are managed is very uniform. Low and intermediate level waste streams are processed in three basic steps:

1. Pre-treatment (if necessary) is applied to adjust the waste properties according to its expected treatment and conditioning. In practice, adjustment of acidity is most commonly used.
2. Treatment is applied to reduce the volume of 'as generated' waste for further conditioning. There are two principal approaches: more commonly concentration of primary waste usually by evaporation and less commonly separation of radionuclide contamination either by ion exchange or advanced ultrafiltration techniques. In the first case, the treatment results in a relatively small volume of waste concentrate with high salinity. In the second case, radionuclides are concentrated in a special ion exchange column or in the filtration material. In both cases, liquid concentrate, or spent filtration materials and filters are further addressed for final conditioning. Bulk condensate from evaporation or filtrate from ion exchange or filtration procedure can be, after proper control, reused or even cleared for discharge.

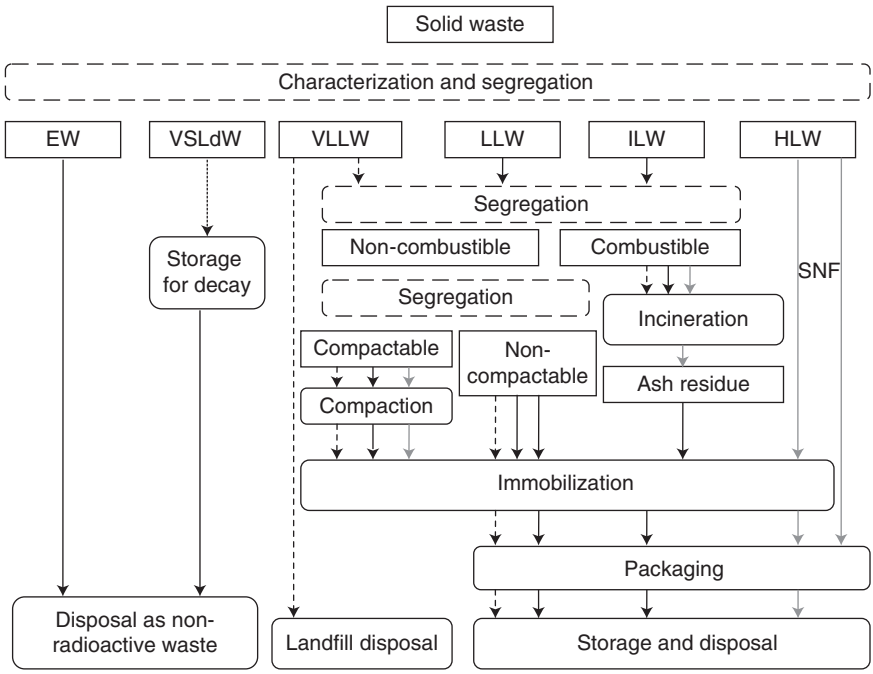


2.1 Principal scheme of liquid RAW management [12].

3. Conditioning of liquid waste concentrates is the process of their incorporation into a selected matrix. The resulting waste form is loaded into a proper container. Cement and bitumen are commonly used matrices; polymer, geopolymer and some other matrices are less frequently encountered. Recently, also cold crucible vitrification has been considered for concentrate conditioning, resulting in an excellent waste form. Selection of the container usually depends on the waste form properties, the national waste management policy, the selected disposal option and the waste acceptance requirements.

The principal scheme of solid RAW management is presented in Fig. 2.2, taken from Ref. [12]. The composition and properties of 'as generated' solid radioactive waste are substantially more variable than liquid waste. Therefore also the technologies used for processing solid waste are more variable and should be tailored according to the individual requirements and expectations of the waste generator. Similar to liquid waste, low and intermediate level solid waste streams are processed in three basic steps:

1. Pre-treatment comprises mostly sorting and segregation of the waste according to waste characteristics and the expected processing



2.2 Principal scheme of solid RAW management [12].

technologies. An extremely important feature of the pre-treatment step is a chance to segregate non-active waste from the bulk waste and contribute in this way to the minimization of waste generation. The potential for reduction of solid waste generation is considerably higher than that of liquid waste. Given the present state of technology, the best results in volume minimization can be achieved with a combination of thermal treatment (pyrolysis) and compaction technology. Therefore, the most common approach is to segregate the waste into combustible and non-combustible categories and the non-combustible category is further segregated into compactable and non-compactable waste. Metallic waste represents a special category of solid waste and its processing is a separate issue. Decontamination is often used as a pre-treatment or treatment procedure in this case.

2. Treatment steps are applied to reduce the waste volume. Combustible waste can be thermally treated by pyrolysis with follow-up conditioning of ash by incorporation into a matrix or by supercompaction. In some countries there are objections to the application of the high temperature thermal treatment resulting in generation of exhaust gases. In this case, compaction or medium temperature thermal destruction in the absence

of air (steam reforming) are the only applicable options for volume reduction of solid waste. However, steam reforming is still not commonly used in RAW management. There are two principal options for compaction of the radioactive waste: low pressure in drum compaction and, more favourable for operational waste, high pressure compaction (supercompaction), when the whole drum filled with waste is compacted. Pellets are obtained in this conditioning step and can be placed into a container and/or encapsulated in a proper matrix.

3. Conditioning of solid waste is in principle immobilization of treated waste into a proper matrix, most commonly cement grout. Ash from a high temperature thermal treatment facility is either directly cemented in a container, or first compacted and the pellets from the compactor are encapsulated (grout or another matrix) into a proper container. Non-compactable waste is adjusted and sorted by size and usually encapsulated (e.g., grouted) in a proper container. In some cases, non-compactable waste is packed into drums and the filled drums are then loaded into special containers (e.g., reinforced concrete containers) and the void space is filled with cement grout. In all the above-mentioned cases, cement grout can be prepared with liquid radioactive waste to provide better utilization of container space.

Each waste processing technology uses specific types of containers for accommodation of waste forms. The most common container worldwide is the standard 200 L metallic drum, made from various types of steel (in general carbon steel and/or stainless steel) with consideration of various corrosion protection measures. Waste packages made of 200 L drums are commonly accepted for final disposal. However, in some countries there is a requirement to place 200 L drums with conditioned waste into special reinforced concrete containers and fill the void space by cement grout or another appropriate filling material. Since reinforced concrete containers themselves provide a 300-year leak-tightness guarantee, such an approach can be considered as an important contribution to the long-term safety of waste disposal. This is of special importance in densely populated countries, where disposal facilities are located close to settled sites.

Spent ion exchange resins and sludge, sometimes generated during waste evaporation or formed as sediment at the bottom of storage tanks, are a special waste category, which usually causes some processing problems. Direct incorporation of the resins/sludges/sediments into most common cement matrices requires a special procedure and modifications of the cement matrix which sometimes leads to a waste form with insufficient mechanical and durability properties. There are some other options: application of another matrix, compatible with the organic structure of spent

resins (e.g., polymers or geopolymers), high temperature pyrolysis, medium temperature destruction (steam reforming), etc. Another option is to pack spent resins and sludge into special high integrity containers and consider their long-term storage or disposal in underground repositories or deep geological formations.

As has been demonstrated in this section, the selection of the individual steps and technological sequences involved in RAW processing is a serious problem with many various aspects to be considered. Each waste stream requires an individual approach and an individual selection of the proper waste encapsulation matrix, waste form and waste container. In this chapter, therefore, only general considerations have been presented that should be taken into account in the waste management planning process.

2.5 Sources of further information

This section is a general introduction to RAW management planning and implementation. The objective is to provide a basic orientation with inputs as to what should be considered during waste processing planning and selection of the proper waste processing technological sequences. More detailed technical and technological information can be found in other chapters of this book, dedicated to waste processing technologies.

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3

International safety standards for radioactive waste (RAW) management and remediation of contaminated sites

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Abstract: This chapter presents the key factors and current status of the development of international safety standards and recommendations on safe remediation and radioactive waste management. It highlights the international legally binding instruments, safety fundamentals, safety requirements and guides, with specific emphasis on pre-disposal and disposal of radioactive waste, as well as transport.

Key words: safety, standards, pre-disposal, disposal, radioactive waste, transport, remediation.

3.1 Introduction

3.1.1 Radiation safety and radioactive waste (RAW)

Safety is the prime consideration during management of RAW on account of the potential that exists for exposure of people to radiation. Such exposure can be to workers involved in the handling and management of RAW or to members of the public, due to any radioactive material associated with the waste being released into the publicly accessible environment. Similarly, environments contaminated with radioactive materials can cause exposure of persons to radiation. Both the management of RAW and contaminated environments can also lead to plant and animal species being exposed to radiation. Exposure can arise at the present time and can also occur in the future, and its magnitude can vary from insignificant to very high depending on the nature of the RAW and the circumstances of exposure. Exposure can also arise during normal anticipated circumstances associated with waste management and contaminated environments and from accidents or disruptive events.

The same philosophical basis for radiation safety has been adopted for all facilities and activities that can give rise to radiation exposure. However,

the manner in which this philosophical basis has been developed and applied to waste management and contaminated environments is influenced by the often long timescales involved and the desire to dispose of the waste, i.e. to no longer have to exercise active control and management over the materials. There is also need to differentiate those materials containing radioactive material but at such low levels that the material does not need to be managed as radioactive waste.

The effects of exposure to radiation have been studied throughout the twentieth century and a sound knowledge base has been developed [1]. Studies continue to refine and update this knowledge base, but in general the effects are known. Lower levels of radiation dose cause an increase in the incidence of cancer in the exposed populations and at higher levels of radiation dose in excess of a threshold in the region of 1 Gy, deterministic health impacts start to occur. The latter effects range from chromosomal aberrations to organ damage and skin burns to death at doses beyond a few Gy. The rate of cancer incidence increases with increasing radiation dose in a stochastic manner; in the lower range of doses, no increase in the natural incidence of cancers is detectable, at higher levels of dose in larger populations, an excess incidence is discernible. The basic approach to radiation safety is both to prevent short-term deterministic health effects and to ensure that the longer-term risk of cancer induction is not significant.

Exposure to radiation can arise from radioactive material emitting penetrating radiation located outside the body of a person or other species, which due to proximity impinges on the body. Alternatively, exposure can arise from radioactive material being incorporated into the body, generally by inhalation or ingestion. Other diffusive transfer mechanisms generally apply to incorporation into plants.

All these factors have to be considered in developing and applying a safety regime for the management of RAW and contaminated environments. The fundamental approach is to reduce the volume of waste to the extent reasonably possible, to solidify it into an immobile form, and to provide measures to contain and isolate the waste from the accessible environment. The containment is intended to keep the radionuclides within the containment boundary by chemical or physical fixation within the waste matrix and by physical containing barriers, and to provide shielding for any penetrating radiation emitted from the radionuclides within the waste [2]. The isolation function aims to keep the radionuclides away from people and the environment and also to protect the waste and its protective features from disturbing and degrading influences such as fire, water, physical disruption, etc. The timeframes required for such containment and isolation are influenced by the radioactive half-lives of the radionuclides contained in the waste.

Many activities involving radioactive material processing, handling and use also give rise to contaminated effluents. Treatment of the fluids

generally involves cleaning by filtration, solvent extraction, ion exchange or by evaporation. The aim of these cleaning processes is to reduce the radioactivity levels in the effluent to the extent that they can be safely discharged into the environment.

This chapter outlines the general international principles of radiation, waste and transport safety. Examples of their application in various countries are given in Part II.

3.1.2 Types and origins of RAW

Radioactive waste can arise from a number of activities and facilities. It can occur in a very broad range of physical and chemical forms, and can have a similarly wide range of associated radiological properties. These factors influence the possible mechanisms of radiation exposure to persons and other species and the potential magnitude of such exposure.

One of the major sources of RAW generation is the nuclear sector, including both the commercial nuclear power industry and the military nuclear weapons manufacturing component. Whilst having completely different objectives, the waste types generated have many similarities, arising from uranium mining and processing, enrichment, nuclear fuel manufacture, reactor operation, reprocessing and decommissioning. Production and use of radioactive sources for industrial, medical and other applications is another significant source of radioactive waste generation. The sources can be reactor produced, so the waste types have some similarities to the nuclear sector, or can be accelerator produced. Radioactive sources are generally of small physical size (i.e. < centimetres) but can vary significantly in terms of radiological properties – half-life, radioactive content and radiation type emitted. Radiation sources are widely used in medicine, industry and research. A number of scientific research and development activities use or generate radioactive material and can give rise to a broad and diverse range of RAW. The other area in which RAW arises is that involving naturally occurring radionuclides, generally associated with mineral extraction and processing. Numerous ore bodies and mineral deposits contain elevated levels of naturally occurring radionuclides, often linked to the phosphate industry, coal mining and oil extraction. Water treatment for domestic use can give rise to sludges with concentrations of naturally occurring radionuclides that warrant management as radioactive waste.

As indicated, RAW can take many different forms, a factor influencing safety and hence the way in which the waste is managed. The waste material itself can be radioactive, it can contain radioactive material or it can be contaminated on its surfaces by radioactive material. A considerable amount of waste is generated in the form of solids, varying from granular mineral forms to solid rock to civil rubble to equipment, metals, plastic and

paper. Also, contaminated liquids and gases are generated whose treatment can give rise to solid waste such as ion exchange resins, cemented or bitumenised chemical sludges and filters used to clean liquid or gas streams contaminated with particulate and volatile radioactive species.

The majority of nuclear activities commence with mining and processing uranium/thorium-bearing ores, likewise such waste can arise from other mining and mineral processing activities. The radioactive species contained in these ores originate from the primordial radionuclides with radioactive half-lives of the order of thousands of millions of years. These species, isotopes of uranium and thorium, each head decay chains of radionuclides with radioactive half-lives varying from microseconds to thousands of years. A decay product of particular interest is radon, the radioactive noble gas whose physical characteristics influence its instant mobility and related radiological hazard potential. Like any mining and mineral processing activity, the residues are waste rock, process tailings, chemical sludges and used plant equipment and buildings. Many process fluids are used, as mines often have to be de-watered and both mines and processing buildings are normally ventilated. Thus the spectrum of physical waste types generated takes the form of solids, liquids and gases. The amounts of waste generated are large; hundreds to thousands of tonnes of rock are mined to produce a single tonne of uranium. Mine sites are also generally quite large (i.e. up to tens of square kilometres) in area and due to the bulk nature of the materials handled, stored and processed, large areas of land and buildings become radioactively contaminated during operations. On the other hand, the radioactive concentration of the materials involved is not high – on the order of becquerels per kilogramme, although various adventitious concentration mechanisms can cause these concentrations to multiply thousands of times.

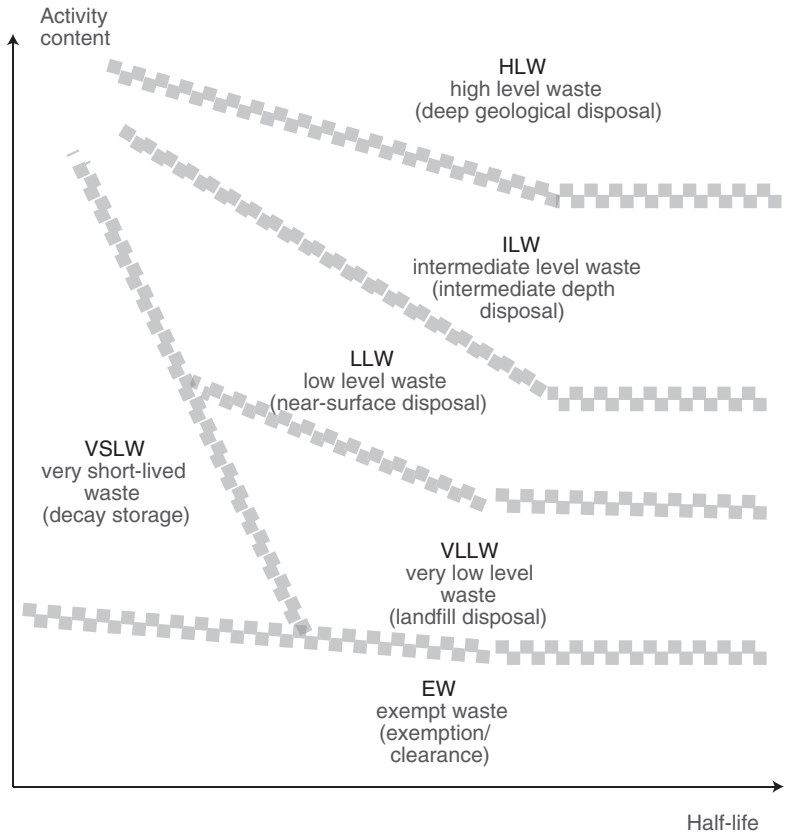
3.1.3 RAW classifications

Radioactive waste can be classified according to different schemes for different purposes such as operational segregation and treatment, e.g. compressible waste and combustible waste. At an international level, an agreed system of classification has been developed based on long-term safety considerations [3]. The scheme is used at a national policy and strategy level for exchange of information and for the purposes of international safety standards and international safety conventions. The scheme is based on linking waste types to corresponding disposal options. The waste classes defined are summarised in Box 3.1.

The classification scheme is illustrated graphically in Fig. 3.1. The ordinate is the radioactivity content of the waste and the abscissa the half-life. The diagram illustrates the need for greater levels of containment and isolation for higher activity and longer lived radioactive waste.

Box 3.1 IAEA classification of radioactive waste

(1) **Exempt waste (EW):** Waste that meets the criteria for clearance, exemption or exclusion from regulatory control for radiation protection purposes.
(2) **Very short lived waste (VSLW):** Waste that can be stored for decay over a limited period of up to a few years and subsequently cleared from regulatory control according to arrangements approved by the regulatory body, for uncontrolled disposal, use or discharge. This class includes waste containing primarily radionuclides with very short half-lives often used for research and medical purposes.
(3) **Very low level waste (VLLW):** Waste that does not necessarily meet the criteria of EW, but that does not need a high level of containment and isolation and, therefore, is suitable for disposal in near-surface landfill-type facilities with limited regulatory control. Such landfill-type facilities may also contain other hazardous waste. Typical waste in this class includes soil and rubble with low levels of activity concentration. Concentrations of longer lived radionuclides in VLLW are generally very limited.
(4) **Low level waste (LLW):** Waste that is above clearance levels, but with limited amounts of long-lived radionuclides. Such waste requires robust isolation and containment for periods of up to a few hundred years and is suitable for disposal in engineered near-surface facilities. This class covers a very broad range of waste. LLW may include short-lived radionuclides at higher levels of activity concentration, and also long-lived radionuclides, but only at relatively low levels of activity concentration.
(5) **Intermediate level waste (ILW):** Waste that, because of its content, particularly of long-lived radionuclides, requires a greater degree of containment and isolation than that provided by near-surface disposal. However, ILW needs no provision, or only limited provision, for heat dissipation during its storage and disposal. ILW may contain long-lived radionuclides, in particular, alpha-emitting radionuclides that will not decay to a level of activity concentration acceptable for near-surface disposal during the time for which institutional controls can be relied upon. Therefore, waste in this class requires disposal at greater depths, of the order of tens of metres to a few hundred metres.
(6) **High level waste (HLW):** Waste with levels of activity concentration high enough to generate significant quantities of heat by the radioactive decay process or waste with large amounts of long-lived radionuclides that need to be considered in the design of a disposal facility for such waste. Disposal in deep, stable geological formations usually several hundred metres or more below the surface is the generally recognised option for disposal of HLW.



3.1 Radioactive waste classification [3].

3.1.4 Global inventories of RAW

Radioactive waste has been generated in increasingly large amounts since the advent of the nuclear industry in the 1940s and 1950s. Contaminated environments have also been experienced from that era; a considerable number since the start of uranium mining, some from military-related activities – both from processing plants associated with weapons production and weapons testing and from nuclear accidents, in particular Chernobyl and more recently Fukushima. The amount of RAW generated to date in non-military programmes is generally reported in the open literature, but that from military activities can only be estimated from weapons production activities. A review of the global inventory has been made by the IAEA and is summarised in Table 3.1 [4].

Table 3.1 Cumulative global radioactive waste inventory

Waste source	Low- and intermediate-level waste (LILW)		Spent fuel[a]		HLW[b]		Mining & milling	
	Volume (m^3)	Activity (TBq)	Mass (MTHM)	Activity (TBq)	Volume (m^3)	Activity (TBq)	Volume (m^3)	Activity (TBq)
Nuclear fuel cycle	2.2 E6	1.2 E6	1.8 E5	2.8 E10	3.4 E4	4.2 E7	1.6 E9	2.8 E4
Institutional activities	1.1 E6	7.0 E5						
Defence and weapon	4.0 E6	7.0 E5			8 E5[c]	3.1 E7[c]	2.5 E8	4.6 E3
Total	7.3 E6	2.6 E6	1.8 E5	2.8 E10	8.3 E5	7.3 E7	1.8 E9	3.3 E4

[a] In reality a relatively minor fraction of the spent fuel generated by nuclear power plants (NPPs) has been reprocessed and has been transformed into a variety of products, including different classes of radioactive waste.
[b] A fraction of the HLW generated by reprocessing civilian spent fuel has been vitrified. Most HLW generated by defence programmes is stored in liquid form.
[c] Estimates are highly uncertain. In some countries there is no clear separation between reprocessing for military and for civilian purposes.

3.2 International, regional and national organisations involved

A number of international organisations play a key role in establishing the basis for radiation protection and its implementation in international safety standards, which are widely adopted in national legal and regulatory frameworks. The main organisations are identified in this section.

3.2.1 The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR)

The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) was established by the General Assembly of the United Nations in 1955 to assess and report levels and effects of exposure to ionising radiation [1]. Initially its work was based on the consequences of the Hiroshima and Nagasaki bombings in Japan. Later, scientific input from the UN member countries has provided the basis for development of international recommendations for protection of humans and the environment (carried out by the International Commission on Radiological Protection (ICRP), see below). The UNSCEAR secretariat collates relevant data submitted by the member countries, international organisations and non-governmental organisations, and analyses those data prior to publication of the scientific evaluations (e.g., assessment of the consequences of the Chernobyl accident [5]). Governments and organisations throughout the world rely on the Committee's[1] estimates as the scientific basis for evaluating radiation risk and for establishing protective measures. The latest report of UNSCEAR [6] of 2011 focuses on summarising low-dose radiation effects on health.

3.2.2 The International Commission on Radiological Protection (ICRP)

The International Commission on Radiological Protection (ICRP) was established in 1928 and since then has developed, maintained and elaborated the International System of Radiological Protection used worldwide as the common basis for radiological protection standards, legislation, guidelines, programmes and practice. At present the ICRP recommendations are based on the input from UNSCEAR, the current understanding of scientific data and also taking into account the societal and ethical aspects.

[1]At present the UNSCEAR committee comprises 27 countries (Argentina, Australia, Belarus, Belgium, Brazil, Canada, China, Egypt, Finland, France, Germany, India, Indonesia, Japan, Mexico, Pakistan, Peru, Poland, South Korea, Russia, Slovakia, Spain, Sudan, Sweden, Ukraine, United Kingdom, United States of America).

The latest revised Recommendations for a System of Radiological Protection 2007 [7] formally replaced the Commission's previous, 1990, Recommendations [8]; and update, consolidate and develop the additional guidance on the control of exposure from radiation sources issued since 1990.

ICRP is an independent international organisation with more than 200 volunteer members (leading scientists and policy makers in the field of radiological protection) from approximately 30 countries [9].

3.2.3 The International Atomic Energy Agency (IAEA)

According to its 1956 statute [10], the IAEA together with its Member States[2] has the mandate to develop international safety standards (on nuclear, waste, radiation and transport safety) and to provide for their application by regulators, operators, technical support organisations and vendors. The standards on waste safety are made up of safety fundamentals, safety requirements and safety guides (see Section 3.4) that address safety criteria, and best practices for compliance during generation, management prior to disposal and disposal of all types of RAW. The IAEA applies various mechanisms to assist Member States in applying the safety standards, such as peer reviews (pre-disposal [11] and disposal [12]), technical cooperation projects [13]; international inter-comparison projects (ISAM [14], ASAM/PRISM [15], SADRWMS [16], etc.); training and education, and exchange of information, e.g. conferences [17, 18] and thematic workshops [19].

3.2.4 The Nuclear Energy Agency (NEA) of the Organisation for Economic Co-operation and Development (OECD)

The Nuclear Energy Agency (NEA) of the Organisation for Economic Co-operation and Development (OECD) is an international organisation of 31 Members[3] with the mission to assist its member countries in maintaining and further developing, through international co-operation, the scientific, technological and legal bases required for the safe, environmentally friendly and economical use of nuclear energy for peaceful purposes [20]. Radioactive waste management is one of the areas of NEA's work which includes:

[2] 159 Member States (as of February 2013).

[3] In February 2013: Australia, Austria, Belgium, Canada, Czech Rep., Denmark, Finland, France, Germany, Greece, Hungary, Iceland, Ireland, Italy, Japan, Luxemburg, Mexico, Netherlands, Norway, Poland, Portugal, Rep. of Korea, Russian Federation, Slovak Rep., Slovenia, Spain, Sweden, Switzerland, Turkey, United Kingdom and the United States of America.

nuclear safety and regulation, nuclear energy development, radiological protection and public health, nuclear law and liability, nuclear science, maintenance of data banks and information and communication. With respect to RAW management, the NEA focuses on providing assistance to member countries in developing safe, sustainable and societally acceptable strategies for management of all types of radioactive materials, with particular emphasis on the management of long-lived waste and spent fuel and on decommissioning of nuclear facilities.

3.2.5 Other organisations and programmes

Other UN programmes, such as the United Nations Environmental Programme (UNEP) and the United Nations Development Programme (UNDP) [21] implement a number of initiatives aimed at remediation of contaminated sites in Central and East Asia. One of these projects is the ENVSEC Initiative (Environment and Security Initiative) 'Strengthening Coordination of Project Formulation and Mobilization of Resource for Sustainable Radioactive Waste Management in Central Asia'. The latter is targeting contaminated sites with uranium tailings in the Kyrgyz Republic, Republic of Kazakhstan, Republic of Tajikistan and Republic of Uzbekistan.

In addition to the above international organisations, regional organisations and other groups are involved in initiatives concerning the regulation of nuclear, waste and radiation safety, e.g.:

- The European Commission (EC) establishes policies, directives, regulations and recommendations in the field of nuclear energy, including the safe management of spent fuel and radioactive waste. In 2007, following a decision of the EC, the European Nuclear Safety Regulators Group (ENSREG) was established as an independent, authoritative expert body. Its aim is to help to establish the conditions for continuous improvement and to reach a common understanding in the areas of nuclear safety and radioactive waste management. It is composed of senior officials from the national nuclear safety, RAW safety or radiation protection regulatory authorities from all 27 Member States in the European Union and representatives of the EC [22]. Recently the European Commission approved a new Directive on the management of spent fuel and radioactive waste [23].
- Regulatory associations (networks) in Africa (Forum of Regulatory Bodies in Africa – FNRBA [24]), Europe (WENRA – Western European Nuclear Regulators Association [25]), Latin America (Latin American Forum of Nuclear and Radiological Regulatory Organisations – FORO [26]), Arab countries – ANNuR (Arab Network for Nuclear Regulators [27]), Asia – Asian Nuclear Safety Network (ANSN

[28]) were established, as well as a Regulatory Cooperation Forum (RCF) for new countries entering into nuclear energy [29], etc.

At a national level, the different organisations that play a key role in the establishment and implementation of systems for management and control of radioactive waste can be summarised as follows:

- operators (such as NPPs, research facilities, medical laboratories) that produce and handle waste and their technical support organisations (TSOs, e.g., contractors);
- regulatory authorities that perform regulatory oversight during all steps of RAW management (it is also possible that these authorities use the services of dedicated TSOs);
- in some countries a dedicated radioactive waste management organisation is established to deal with the pre-disposal management (e.g., COVRA in the Netherlands [30]), long-term management (i.e., storage and disposal) of this waste (e.g., ANDRA in France [31]; SE 'RAO' in Bulgaria [32], PURAM in Hungary [33], NWMO in Canada [34]) or even decommissioning of nuclear facilities (ENRESA, Spain [35] and NDA, UK [36]);
- financial authorities that in many cases control the national funds for radioactive waste management and decommissioning;
- research institutions that are involved in supporting research activities such as site investigation and understanding of the phenomena influencing safety.

3.3 International standards for radiological safety and environmental protection

As indicated previously, the approach to radiation safety that has been accepted universally is to prevent deterministic effects (acute effects such as skin damage, organ damage and ultimately death) and reduce the likelihood of stochastic effects (cancer and genetic effects) to levels that are not significant. The underlying concepts to applying this approach are elaborated extensively in the recommendations of the ICRP, an international commission established in 1928 by the International Congress on Radiology [9]. This was on account of the increasing recognition of the dangers associated with medical uses of X-rays by the medical profession and the need to establish appropriate protection regimes. The work of the ICRP increased substantially in the second half of the twentieth century with the advent of the nuclear industry and the legacy of nuclear weapons development and use towards the end of the Second World War. The ICRP has updated its recommendations on a regular basis around every ten years as knowledge about the effects of exposure to radiation has increased and as

the philosophy of protection has developed and matured. The ICRP is an independent scientific body and its recommendations have no legal standing. The recommendations are, however, generally used by the international standards setting bodies as a basis for developing international standards. With regard to radiation safety, the IAEA is the body mandated by the United Nations to establish international safety standards for nuclear, radiation, RAW and radioactive material transport safety. The safety standards are based on a set of fundamental safety principles [37] endorsed both technically and politically by all the member countries of the IAEA. The principles are set down in Box 3.2.

The system of radiation protection recommended by the ICRP and adopted in the international safety standards for radiation protection of the IAEA[4] [38] has three general principles, which concern justification of practices and activities, optimisation of protection and the application of individual radiation dose limits, and are expressed in Safety Principles 4, 5, 6 and 10 in Box 3.2. The system of protection also has a number of dimensions; it differentiates three types of exposure situations, namely planned, emergency and existing situations and three types of exposure: occupational, public and medical. The first of these three, 'occupational', concerns exposure of persons on account of their work situation, such as workers in a nuclear power plant, a waste management facility, a uranium mine or a nuclear medicine department in a hospital. 'Medical exposure' is exposure of patients to radiation for either diagnostic (e.g., X-ray) or therapeutic (e.g., radiotherapy) purposes. Medical exposure is not relevant to RAW management or contaminated areas and will not be dealt with further. The third category is 'public exposure' and is all exposures other than occupational or medical. Public exposure can arise from various causes such as discharge into the environment of radioactive effluents, migration of radionuclides from radioactive waste disposal facilities into the accessible biosphere, transport of radioactive material in public areas, the use of radioactive consumer products, etc.

Planned situations are those in which a facility or activity where radioactive material will be present is knowingly and deliberately developed and operated. This means that safety and protection measures can be planned and put in place, and the radiation exposure of people associated with the activity or operation of the facility can be controlled to whatever level is decided. The protective measures can be design features of the facility such as shielding or containment features or can be operational controls such as limiting access in certain areas or decontaminating working areas. All

[4]The standard is entitled 'Radiation Protection and Safety of Radiation Sources – International Basic Safety Standards', but is commonly referred to as the Basic Safety Standards or BSS.

Box 3.2 International fundamental safety principles

Principle 1: Responsibility for safety
The prime responsibility for safety must rest with the person or organisation responsible for facilities and activities that give rise to radiation risks.

Principle 2: Role of government
An effective legal and governmental framework for safety, including an independent regulatory body, must be established and sustained.

Principle 3: Leadership and management for safety
Effective leadership and management for safety must be established and sustained in organisations concerned with, and facilities and activities that give rise to, radiation risks.

Principle 4: Justification of facilities and activities
Facilities and activities that give rise to radiation risks must yield an overall benefit.

Principle 5: Optimisation of protection
Protection must be optimised to provide the highest level of safety that can reasonably be achieved.

Principle 6: Limitation of risks to individuals
Measures for controlling radiation risks must ensure that no individual bears an unacceptable risk of harm.

Principle 7: Protection of present and future generations
People and the environment, present and future, must be protected against radiation risks.

Principle 8: Prevention of accidents
All practical efforts must be made to prevent and mitigate nuclear or radiation accidents.

Principle 9: Emergency preparedness and response
Arrangements must be made for emergency preparedness and response for nuclear or radiation incidents.

Principle 10: Protective actions to reduce existing or unregulated radiation risks
Protective actions to reduce existing or unregulated radiation risks must be justified and optimised.

waste management operations are planned (even if the waste is generated by an accident or arose in a previously uncontrolled activity), as are all activities to clean up contaminated areas or decisions not to clean up areas. As such the radiation safety recommendations pertaining to planned situations will apply. Despite measures to prevent accidents and the fact that such accidents are rare, they can happen during RAW management

activities and for such circumstances the recommendations for emergency situations would apply. Unlike planned exposure situations, it is not possible to control exposures from accidents *a priori* and as such the requirements are in general to plan for accidents and emergencies and to put mitigatory measures in place to limit exposures to the extent reasonably possible in the event of an accident. Contaminated environments do exist on account of accidents such as Chernobyl or past uncontrolled practices such as some uranium mining activities, and these are considered existing exposure situations. In existing exposure situations, exposures are occurring and the system of protection calls for measures to be put in place to reduce exposures such as limiting access to certain areas or cleaning up of contaminated facilities and/or land.

3.3.1 Radiation protection requirements for planned situations

The 'Basic Safety Standards' set down a number of general requirements for planned situations together with specific requirements for the control of occupational and public exposure. The requirements of particular importance to waste management and contaminated environments are elaborated below.

The general requirements call for a graded approach to safety with the safety provisions put in place being commensurate with the magnitude and likelihood of exposure and also for all activities and facilities involving RAW or contaminated land to be subject to regulatory control. Of particular importance for RAW management are the concepts of exemption and clearance, whereby governments are required to identify those activities and facilities that need to fall within the scope of regulations, and those materials or objects that can be exempted from or cleared from control on account of the small amount of radioactivity associated with them and their insignificance from a radiation safety perspective. Criteria for exemption and clearance are that exposures associated with the material or activity should not cause radiation doses in excess of the order 10 μSv or less per year. For very unlikely scenarios, the criterion is 1 mSv. Values have been derived for radioactivity concentrations and total activity amounts in limited quantities of material to be cleared and these are provided in a schedule to the BSS together with a table of activity concentrations for clearing unlimited amounts of material.

Facilities and activities have to be justified, i.e. there must be greater benefit than overall detriment (risk from radiation exposure and costs), protection and safety is to be optimised and radiation doses to individuals are to be limited. The optimisation of protection requires radiation doses and risks to be maintained as low as reasonably achievable (ALARA) and

the individual dose limitation requirements to ensure no individuals are subject to unacceptable individual risk from the activity or facility. The safety of facilities is to be assessed and demonstrated to be acceptable before designs are fixed and facilities constructed and operated. The assessment must take into consideration both normally anticipated circumstances of operation and possible accidents. This forms part of the regulatory process whereby the prospective operator conducts the safety assessment and submits this as part of the safety case supporting the application for authorisation. The regulatory authority evaluates the adequacy of the assessment and if necessary design or operational changes are made as necessary prior to authorisation. Conditions of authorisation are based generally on the assessment, and aim to ensure that assumptions and commitments made in the assessment and licence application are fulfilled in practice. Monitoring provisions are required for active demonstration of compliance with conditions of authorisation, together with cognisance being given to operational feedback.

The control over occupational exposure associated with RAW management facilities and activities and contaminated environments is the same as for any planned activity or facility where radioactive material is present. Requirements include the provision of monitoring and recording of individual radiation doses received, arrangements and programmes to ensure that radiation exposures and risks of accidental exposure are maintained as low as reasonably achievable, i.e. optimisation of safety and protection, monitoring of workers health and training of workers in radiation protection measures. Operational radiation protection programmes must be in place, i.e. radiation hazard areas designated, work rules in place, prior planning of work to be undertaken in radiation areas and monitoring of the working environment.

Public exposure is of particular relevance in respect of waste management and contaminated environments because of the potentially long timeframes involved and potential impacts on multiple generations and the importance of maintaining a sustainable environment for the long-term future. In this regard, assessing the impact of facilities and activities both on current and future generations assumes great importance as part of the overall development of the safety case – the collection of safety arguments and supporting evidence that demonstrate safety. Specific requirements relate to the control over public exposure and concern various parties. Governments must clearly assign responsibilities and must provide a clear regulatory framework. The regulatory framework must provide for continuity in control of materials movement from one party to another and clear regulations on radiation dose limitations must be laid down. In particular, dose constraints that will be applied to individual facilities and activities and demonstration that the requirements to optimise protection will be

fulfilled. If it is intended to discharge effluents to the environment, specific authorisations must be given setting down the amounts and concentrations of radioactivity that are allowed in the discharge and the manner in which this must be monitored and controlled and what reports are to be submitted to the regulatory authority regarding ongoing compliance with the discharge authorisation, together with any aspects of non-compliance.

3.3.2 Radiation protection requirements for existing situations

As with planned situations, governments must ensure that existing exposure situations that have been identified are evaluated to determine which occupational exposures and public exposures are of concern from the point of view of radiation protection. The government and the regulatory body or other relevant authority must ensure that remedial actions and protective actions are justified and that protection and safety is optimised.

Governments need to ensure that provision is made for identifying those persons or organisations responsible for areas with residual radioactive material, for establishing and implementing remediation programmes and post-remediation control measures, if appropriate, and for putting in place an appropriate strategy for RAW management.

3.3.3 Safety requirements for the management of RAW

The international safety standards for RAW management are grouped into those relating to pre-disposal management of RAW [39], disposal of RAW [40], decommissioning of facilities [41] and remediation of contaminated buildings and areas [42]. These are elaborated in the sections below. The safety requirements set out in the standards are aimed at governments, regulators and operator organisations carrying out waste management activities and those parties responsible for contaminated environments.

3.3.4 Safety requirements for pre-disposal management of RAW

The first few requirements are directed at governments and require appropriate national legal and regulatory frameworks to be established within which RAW management activities can be planned and safely carried out. This includes the clear and unequivocal allocation of responsibilities, the securing of financial and other resources, and the provision of independent regulatory functions. Consideration also has to be given to providing protection beyond national borders as appropriate and necessary for neighbouring countries that may be affected. Governments must also ensure that

a national policy and a strategy for RAW management are established that are appropriate for the nature and amounts of RAW in the country. They must indicate the regulatory control required for particular RAW management facilities and activities, and be compatible with any regional or international conventions and codes that have been ratified by the country. The national policy on radioactive waste management and strategy to implement it must then form the basis for decision making with respect to the management of RAW within the country.

The regulatory body needs to establish regulations for the development of RAW management facilities and activities and to set out procedures for meeting requirements for the various stages of the licensing process. It has to review and assess the safety case for RAW management facilities and activities prepared by the operator both prior to authorisation and periodically during operation. Provisions must also be in place for issuing, amending, suspending or revoking licences, subject to any necessary conditions and the regulatory body has to carry out activities to verify that the operator meets these conditions.

Operator organisations have the prime responsibility for safety and are required to carry out safety assessments and develop a safety case demonstrating safety. They must also ensure that the necessary activities for siting, design, construction, commissioning, operation, shutdown and decommissioning are carried out in compliance with legal and regulatory requirements. Interdependences among all steps in the pre-disposal management of RAW, as well as the impact of the anticipated disposal option have to be appropriately taken into account and the regulatory authorities must ensure this in the event of different operator organisations having responsibility for different aspects of waste management such as treatment, transport, storage and disposal. An integrated approach must also be taken to both safety and security in the pre-disposal management of RAW. The quality of all work influencing safety must be of a high standard and in this regard appropriate management systems must be applied for all steps and elements of the work undertaken.

All RAW has to be identified and controlled and the amount of RAW arising needs to be kept to the minimum practicable. At various steps in the pre-disposal management of RAW, the RAW has to be characterised and classified in accordance with requirements established or approved by the regulatory body.

All radioactive material for which no further use is foreseen, and with characteristics that make it unsuitable for authorised discharge, authorised use or clearance from regulatory control, has to be processed as radioactive waste. The processing of radioactive waste needs to be based on appropriate consideration of the characteristics of the waste and of the demands imposed by the different steps in its management

(pre-treatment, treatment, conditioning, transport, storage and disposal). Waste packages need to be designed and produced so that the radioactive material is appropriately contained both during normal operation and in accident conditions that could occur in the handling, storage, transport and disposal of waste.

Waste is to be stored in such a manner that it can be inspected, monitored, retrieved and preserved in a condition suitable for its subsequent management with due account taken of the expected period of storage. To the extent possible, passive safety features must be applied in the design and operation of storage facilities. For long-term storage in particular, measures need to be taken to prevent degradation of the waste containment. Waste packages and unpackaged waste that are accepted for processing, storage and/or disposal must conform to criteria that are consistent with the safety case.

The safety case for RAW management facilities and activities is of high importance and operators have to prepare a safety case and a supporting safety assessment, which must also be reviewed and updated from time to time as circumstances evolve. The safety case must include a description of how all the safety aspects of the site, the design, operation, shutdown and decommissioning of the facility, and the managerial controls satisfy the regulatory requirements. It must also demonstrate the level of protection provided and provide assurance to the regulatory body that safety requirements will be met. The safety case and its supporting safety assessment have to be documented at a level of detail and quality sufficient to demonstrate safety, to support the decision at each stage and to allow for independent review and approval. Documentation has to be clearly written and include arguments justifying the approaches taken in the safety case on the basis of information that is traceable.

Waste management facilities must be located and designed so as to ensure safety for the expected operating lifetime under both normal and possible accident conditions, and for their decommissioning. They need to be constructed in accordance with the design as described in the safety case and approved by the regulatory body, and commissioning needs to be carried out to verify that the equipment, structures, systems and components, and the facility as a whole, perform as planned. Facilities have to be operated in accordance with national regulations and with the conditions imposed by the regulatory body. Operations need to be based on documented procedures and due consideration given to the maintenance of the facility to ensure its safe performance. Emergency preparedness and response plans, if required to be developed by the operator, have to be subject to the approval of the regulatory body. Operators have to develop, in the design stage, an initial plan for the shutdown and decommissioning of the predisposal RAW management facility and periodically update it throughout

the operational period. The decommissioning of the facility has to be carried out on the basis of the final decommissioning plan, as approved by the regulatory body. In addition, assurance must be provided that sufficient funds will be available to carry out shutdown and decommissioning.

Some facilities are subject to agreements on nuclear material accounting (nuclear safeguards), and in the design and operation of such facilities the system of accounting for, and control of, nuclear material needs to be implemented in such a way as not to compromise the safety of the facility.

The requirements set out above are aimed at new facilities, but some existing facilities were not developed to such standards and in such cases their safety needs to be reviewed to verify compliance with requirements. Safety related upgrades need to be made by the operator in line with national policies and as required by the regulatory body.

3.3.5 Safety requirements for disposal of RAW

The disposal of radioactive waste is the final step in its management and one aimed at providing a permanent and final safety option. The radiation safety principles and ideas remain the same as for any other aspect of waste management, nevertheless the long timeframes involved give rise to particular challenges which are given particular consideration. The international standards for the disposal of RAW were updated and agreed in 2011 [40] and provide a comprehensive set of safety requirements for all types of waste and disposal options. The standards set down clear safety objectives and criteria (see Box 3.3) and a number of discrete requirements to be fulfilled in order to provide for safety. As with pre-disposal management of RAW, these requirements apply to governments, regulators and operators developing and operating RAW disposal facilities.

Governments are required to establish and maintain an appropriate legal and regulatory framework for safety within which responsibilities are to be clearly allocated for the siting, design, construction, operation and closure of disposal facilities. This must include: confirmation at a national level of the need for disposal facilities of different types; specification of the steps in the development and licensing of facilities of different types; a clear allocation of responsibilities, securing of financial and other resources, and provision of independent regulatory functions relating to planned disposal facilities.

The regulatory body must establish regulatory requirements for the development of different types of disposal facility for radioactive waste and set out the procedures for meeting the requirements for the various stages of the licensing process. It must also set conditions for the development, operation and closure of each individual disposal facility and carry out activities to ensure that the conditions are met.

Box 3.3 Safety objectives and criteria for disposal

Safety objective

The safety objective is to site, design, construct, operate and close a disposal facility so that protection after its closure is optimised, social and economic factors being taken into account. A reasonable assurance also has to be provided that doses and risks to members of the public in the long term will not exceed the dose constraints or risk constraints that were used as design criteria.

Criteria

(a) The dose limit for members of the public for doses from all planned exposure situations is an effective dose of 1 mSv in a year. This and its risk equivalent are considered criteria that are not to be exceeded in the future.
(b) To comply with this dose limit, a disposal facility (considered as a single source) is so designed that the calculated dose or risk to the representative person who might be exposed in the future as a result of possible natural processes affecting the disposal facility does not exceed a dose constraint of 0.3 mSv in a year or a risk constraint of the order of 10^{-5} per year.
(c) In relation to the effects of inadvertent human intrusion after closure, if such intrusion is expected to lead to an annual dose of less than 1 mSv to those living around the site, then efforts to reduce the probability of intrusion or to limit its consequences are not warranted.
(d) If human intrusion were expected to lead to a possible annual dose of more than 20 mSv to those living around the site, then alternative options for waste disposal are to be considered, for example, disposal of the waste below the surface, or separation of the radionuclide content giving rise to the higher dose.
(e) If annual doses in the range 1–20 mSv are indicated, then reasonable efforts are warranted at the stage of development of the facility to reduce the probability of intrusion or to limit its consequences by means of optimisation of the facility's design.
(f) Similar considerations apply where the relevant thresholds for deterministic effects in organs may be exceeded.

Operators of disposal facilities are responsible for the safety of the facilities and must carry out safety assessment and develop and maintain a safety case. They must also carry out all the necessary activities for site selection and evaluation and facility design, construction, operation, closure and, if necessary, surveillance after closure, in accordance with national strategy, in compliance with the regulatory and legal requirements.

The operator of a disposal facility must develop an adequate understanding of the features of the facility and its host environment and of the factors that influence its safety after closure over suitably long time periods, so that a sufficient level of confidence in safety can be achieved. Throughout the

process of development and operation of a disposal facility, an understanding of the relevance and the implications for safety of the available options for the facility must be developed by the operator for the purpose of providing an optimised level of safety in the operational stage and after closure. Operators must evaluate the site and design, construct, operate and close the disposal facility in such a way that safety is ensured by passive means to the fullest extent possible and the need for actions to be taken after closure of the facility is minimised.

The host environment must be selected, the engineered barriers of the disposal facility designed and the facility operated in a manner such as to ensure that safety is provided by means of multiple safety functions, the overall performance of the disposal system not being unduly dependent on a single safety function. Containment and isolation of the waste needs to be provided by means of a number of physical barriers of the disposal system. The performance of these physical barriers must be achieved by means of diverse physical and chemical processes together with various operational controls. In addition, the capability of the individual barriers and controls together with that of the overall disposal system to perform as assumed in the safety case has to be demonstrated.

The engineered barriers, including the waste form and packaging, must be designed, and the host environment selected so as to provide containment of the radionuclides associated with the waste. Containment functions must remain available until radioactive decay has significantly reduced the hazard posed by the waste, and in the case of heat generating waste, containment must be available during the timeframe over which the waste is still producing heat energy in amounts that could adversely affect the performance of the disposal system. Disposal facilities must be sited, designed and operated in such a manner that provides features that are aimed at isolation of the RAW from people and from the accessible biosphere. The features must aim to provide isolation for several hundreds of years for short-lived waste and at least several thousand years for intermediate and high level waste. In providing isolation, consideration needs to be given to both the natural evolution of the disposal system and events causing disturbance to the facility. An appropriate level of surveillance and control has to be applied to protect and preserve the passive safety features, to the extent that this is necessary for them to fulfil the functions that they are assigned in the safety case for safety after closure.

In developing disposal facilities, it is important that a systematic step-by-step process is adopted. Each step must be supported, as necessary, by iterative evaluations of the site, of the options for design, construction, operation and management, and of the performance and safety of the disposal system.

As with pre-disposal facilities and activities, a safety case and supporting safety assessment needs to be prepared and updated by the operator, as

necessary, at each step in the development of a disposal facility, during its operation and after closure. The safety case and supporting safety assessment must be submitted to the regulatory body for approval and must be sufficiently detailed and comprehensive to provide the necessary technical input for the regulatory process and for informing the decisions necessary at each step. The scope of the safety case for a disposal facility must include a description of all safety relevant aspects of the site, the design of the facility and the managerial control measures and regulatory controls that will be applied. It must demonstrate the level of protection that will be provided for people and the environment and provide assurance to the regulatory body and other interested parties that all safety requirements will be met. The safety case and supporting safety assessment have to be documented to a level of detail and quality sufficient to inform and support decisions to be made at each step and to allow for independent review.

The site for a disposal facility must be characterised at a level of detail sufficient to support a general understanding of both the characteristics of the site and how the site will evolve over time. This needs to include its present condition, its probable natural evolution and possible natural events, and also human activities in the vicinity that may affect the safety of the facility over the period of interest. It must also show a specific understanding of the impact on safety of features, events and processes associated with the site and the facility.

The disposal facility and its engineered barriers have to be designed to contain the waste with its associated hazard, to be physically and chemically compatible with the host geological formation and/or surface environment, and to provide safety features after closure that complement those features afforded by the host environment. The facility and its engineered barriers must be designed to provide safety during the operational period. The facility must be constructed in accordance with the design as described in the approved safety case and supporting safety assessment and in such a way as to preserve the safety functions of the host environment that have been shown by the safety case to be important for safety after closure. Construction activities must be carried out in such a way as to ensure safety during the operational period.

Facilities have to be operated in accordance with the conditions of the licence and the relevant regulatory requirements so as to maintain safety during the operational period and in such a manner as to preserve the safety functions assumed in the safety case that are important to safety after closure. At the end of operations, disposal facilities must be closed in a way that provides for those safety functions that have been shown by the safety case to be important after closure. Plans for closure, including the transition from active management of the facility, need to be well defined and practicable, so that closure can be carried out safely at an appropriate time.

Waste packages and unpackaged waste accepted for emplacement in a disposal facility must conform to criteria that are fully consistent with, and are derived from, the safety case for the disposal facility both during operation and after closure. A programme of monitoring needs to be carried out prior to, and during, the construction and operation of a disposal facility and after its closure, if this is part of the safety case. This programme must be designed to collect and update information necessary for the purposes of protection and safety. Information must be obtained to confirm the conditions necessary for the safety of workers and members of the public and protection of the environment during the period of operation of the facility. Monitoring also needs to be carried out to confirm the absence of any conditions that could affect the safety of the facility in the period after closure.

Elements of isolation can be provided by institutional control following the closure of disposal facilities, specifically those on or near to the surface (i.e., a few tens of metres). Plans need to be prepared for the period after closure to address institutional control and the arrangements for maintaining the availability of information on the disposal facility. These plans have to be consistent with passive safety features and must form part of the safety case on which authorisation to close the facility is granted.

In the design and operation of disposal facilities subject to nuclear safeguards, consideration has to be given to ensuring that safety is not compromised by the measures required under the safeguards system. Similarly, measures must be implemented to ensure an integrated approach to safety measures and nuclear security measures.

Management systems to provide for the assurance of quality must be applied to all safety related activities, systems and components throughout all the steps of the development and operation of a disposal facility, the level of assurance for each element being commensurate with its importance to safety.

The safety of existing disposal facilities developed prior to current safety standards needs to be assessed periodically until termination of the licence. During this period, the safety also needs to be assessed when a safety significant modification is planned or in the event of changes with regard to the conditions of the authorisation. In the event that any of the current safety requirements are not met, measures need to be put in place to upgrade the safety of the facility, appropriate economic and social factors being taken into account.

3.3.6 Safety requirements for the decommissioning of nuclear facilities

The main safety requirements for protection of workers, the public and the environment during all stages of decommissioning as set down in the

international safety standards [41] emphasise the importance of considering both radiological and non-radiological hazards in an integrated manner. Decommissioning activities are considered to be part of the original practice, and the safety requirements of the Basic Safety Standards [38] apply to all decommissioning activities.

It is important that a safety culture is fostered and maintained in both the operating organisation and that individuals responsible for decommissioning activities are trained to appropriate levels of awareness of health, safety and environmental matters. Safety needs to be maintained during the entire decommissioning process and beyond if a facility is to be in compliance with the site release criteria [43]. As with pre-disposal and disposal, the safety requirements for decommissioning apply to governments, operator organisations and the regulatory body.

The national legal framework for decommissioning needs to include provisions for the use, possession, storage and handling of all radioactive material generated during decommissioning.

All phases of decommissioning, from the initial plan to the final release of the facility from regulatory control, must be regulated. The regulatory body responsible for all phases of decommissioning must establish the safety standards and requirements for decommissioning, and carry out activities to ensure that the regulatory requirements are met.

The operating organisation is responsible for all aspects of safety and environmental protection during the decommissioning activities and must provide financial assurances and resources to cover the costs associated with safe decommissioning, including management of the resulting radioactive waste.

The operating organisation is also required to define a decommissioning strategy consistent with national decommissioning and waste management policy. The preferred decommissioning strategy is immediate dismantling; however, if another practical strategy is selected it needs to be based on evaluation of factors such as: the availability of waste disposal or long-term storage capacity for decommissioning waste; the availability of a trained workforce; the availability of funds; co-location of other facilities on the same site requiring decommissioning; technical feasibility; and optimisation of the radiation protection of workers, the public and the environment. The strategy must be justified and it must be demonstrated that in the future no undue burdens will be imposed on future generations.

It is important that the strategy includes provisions to ensure that, if final shutdown occurs earlier than expected, the facility shall be brought to a safe configuration and a decommissioning plan is in place for approval and implementation.

It is essential that appropriate means are available to manage waste (including pre-disposal and disposal) of all categories [3] in a timely manner,

with account taken of the overall decommissioning strategy. This involves the application of the concept of clearance [44] of material resulting from decommissioning activities, i.e. material or items released from regulatory control. For sites with more than one facility, a global decommissioning programme needs to be developed for the entire site that ensures interdependences are taken into account in the planning for individual facilities.

It is important that the operating organisation prepares and maintains a decommissioning plan throughout the lifetime of the facility (from the design stage to termination of activities) that shows that the decommissioning can be accomplished safely to meet the defined end state. For existing facilities where a decommissioning plan does not yet exist, a suitable plan for decommissioning needs to be prepared as soon as possible.

The decommissioning plan has to be supported by an appropriate safety assessment covering the planned decommissioning activities and any abnormal events that may occur during decommissioning. The assessment must address occupational exposure and potential releases of radioactive material with resulting exposure of the public.

A graded approach needs to be applied to development of the decommissioning plan commensurate with the type and extent of hazards. The initial plan must be reviewed and updated periodically, at least every five years or as prescribed by the regulatory body, or when specific circumstances warrant, such as if changes in an operational process lead to significant changes to the plan. The plan must address all relevant safety aspects (see [45–47]) such as carrying out a baseline survey of the site, retaining key staff and ensuring that institutional knowledge about the facility is maintained.

Prior to the implementation phase of decommissioning (about 2 years), a final decommissioning plan must be prepared and submitted to the regulatory body for approval. Interested parties have to be provided with an opportunity to review the final decommissioning plan and to provide comments on the plan to the regulatory body prior to its approval.

National legislation must set out the responsibilities with respect to financial provisions for decommissioning (e.g., mechanism for adequate financial resources for safe and timely decommissioning). It is very important that adequate finances for safe decommissioning, including the management of the resulting waste, are available when needed, even in the event of premature shutdown of the facility, and financial assurances to provide for the required resources have to be in place before authorisation to operate the facility is given. If financial assurance for the decommissioning of an existing facility has not yet been obtained, suitable funding provision needs to be put in place as soon as possible. Provision for financial assurance is required prior to licence renewal or extension.

Where the decommissioned facility is released with restrictions on its future use, financial assurance adequate to ensure that all necessary controls

remain effective have also to be obtained before authorisation is terminated by the regulatory body.

An organisation for the management and implementation of decommissioning has to be established as part of the operating organisation, with the responsibility for ensuring that decommissioning will be conducted safely. Regardless of the type of organisational arrangements, the ultimate responsibility for safety remains with the operating organisation, although it is permissible to delegate the performance of specific tasks to a subcontractor. The operator must ensure that individuals responsible for performing activities during the decommissioning process have the necessary skills, expertise and training to complete the decommissioning process safely. This must be in line with a comprehensive quality assurance programme under the operating organisation's management system [48] and be applied to all phases of decommissioning. It is important that the management of the decommissioning project is tailored to the project's complexity and size and to the associated potential hazards.

The operating organisation must implement the decommissioning and related waste management activities in compliance with the national safety standards and requirements. The operating organisation must also inform the regulatory body prior to shutting down the facility permanently and the implementation of the decommissioning plan can only start after regulatory approval is issued.

In the case of deferred dismantling, the operating organisation has to ensure that the facility has been placed, and will be maintained, in a safe configuration and will be appropriately decommissioned in the future. To provide an adequate level of safety, the operating organisation must, *inter alia*, prepare and implement appropriate safety procedures; apply good engineering practice; ensure that staff are properly trained and qualified and are competent; and keep and submit records and reports as required by the regulatory body.

Decontamination and dismantling techniques must be chosen such that the protection of workers, the public and the environment is optimised and the hazards and the generation of waste are minimised.

It is important that prior to using any new or untried decommissioning methods, the use of such methods must be justified and addressed within an optimisation analysis supporting the decommissioning plan. Such analyses must be subject to review and approval by the regulatory body.

Emergency planning arrangements, commensurate with the hazards, need to be established and maintained and incidents significant to safety reported to the regulatory body in a timely manner. A proper waste management path for all waste streams arising from decommissioning activities must also be provided.

Upon completion of decommissioning, it must be demonstrated that the end state criteria as defined in the decommissioning plan and any additional regulatory requirements have been met. The operating organisation can only be relieved of further responsibility for the facility after approval by the regulatory body [43].

A final decommissioning report must be prepared that records, in particular, the end state of the facility or site, and this report must be submitted to the regulatory body for review. In this respect, a system must be established to ensure that all records are maintained in accordance with the records retention requirements of the quality assurance system and the regulatory requirements. If waste remains stored on the site after decommissioning, a revised or new, separate authorisation, including requirements for decommissioning, must be issued for the facility. If a facility cannot be released for unrestricted use, appropriate controls need to be maintained to ensure protection of human health and the environment. These controls must be specified and approved by the regulatory body.

3.3.7 Remediation of contaminated sites and buildings

With regard to remediation of contaminated sites from past practices and/or accidents, a set of international safety requirements has been agreed [42]. When dealing with post-accident situations, it is assumed that all immediate protective actions have already been undertaken.

The objective of remediation is to achieve optimised arrangements for protection of the public, the workers and the environment that maximises the net benefit to society. The goal of remediation activities is the timely and progressive reduction of hazard and eventually, if possible, the unrestricted release of sites. However, there are situations where this goal cannot be achieved in a practical manner and then it must be demonstrated that at least any unacceptable risks to human health and the environment has been removed. Overall remediation activities must be aimed at reducing existing exposures and averting the potential for prolonged exposures to occur in the future.

The risks associated with non-radiological hazards should also be assessed in combination with radiological risks to develop an optimised remediation strategy. In the choice of the optimised remediation option, a wide variety of factors must be considered, such as the impacts on health, safety and the environment; and technical, social and financial factors.

Following the general requirements for radiation protection the remedial measures and protective actions that are to be implemented must be justified and optimised. A generic reference level for aiding decisions on remediation is an existing annual effective dose to the most exposed group of 10 mSv from all sources, including natural background radiation. Nevertheless, remedial

measures would often be justified below the generic reference level and national authorities may define a lower reference level for identifying areas that might warrant remediation in the prevailing circumstances.

For all situations in which the dose thresholds for deterministic effects may be exceeded, the implementation of remedial measures or restrictions on access is required. An existing annual equivalent dose of 100 mSv (inclusive of all existing contributions, including doses due to natural background radiation) to any organ justifies intervention under almost any circumstances, although national authorities could specifically determine that such measures are not justified in exceptional circumstances.

With respect to radiation protection of workers, during the implementation of remedial measures, the exposure of workers must be controlled under the system of radiation protection for practices and the relevant limits must apply [38].

A national remediation strategy is needed to specify, prioritise and to ensure remediation situations are managed in a manner commensurate with the risks associated with the contaminated areas and taking into account possible effects on neighbouring countries. An appropriate waste management strategy and an associated legal framework must be established that are capable of dealing with the waste that arises from the remediation of contaminated areas.

National laws and regulations covering such matters as occupational and public radiation protection, environmental protection, transport of radioactive material, mining of ores and food standards, which may be administered by different government bodies, must be applied in a coherent regulatory process. The legal framework must ensure that adequate funding mechanisms are available and that responsibilities are assigned for the financing of remedial measures and protective actions to be taken after remediation that are proportionate, manageable and economically sustainable.

In formulating the national remediation strategy, it may be necessary to involve a number of government and private organisations, and other stakeholders. Also stakeholders' involvement must be considered in the definition, implementation and verification of remediation programmes, and mechanisms for regular public information exchange on the implementation of these programmes must be in place.

The identified responsible parties for the remediation of an area are responsible for all aspects of safety until completion of the remediation activities. To ensure an adequate level of safety, the responsible parties must perform safety assessments and environmental impact assessments; prepare and implement appropriate safety procedures; apply good engineering practices; ensure that the staff are trained, qualified and competent; establish and implement a quality assurance programme; and keep records as required by the regulatory body.

The responsible parties must also: (i) prepare and maintain remediation plans; (ii) establish and maintain arrangements for emergency planning commensurate with the hazards associated with the remediation activities; (iii) report incidents significant to safety to the regulatory body in a timely manner; (iv) identify an acceptable waste disposal or storage site, as appropriate, for the generated waste; and (v) ensure that all waste is transported safely and in accordance with the requirements for its transport.

In general, remediation of a contaminated area involves preparation and approval of a remediation plan; remediation operations; and management of radioactive waste resulting from the remediation activities. It needs to be based on collection and assessment of all available information of current and past activities at the site. Therefore an appropriate assessment of both the radiological and non-radiological impacts of the situation must be performed and the benefits and detriments associated with possible remedial measures, including the associated restrictions and institutional arrangements following remediation must be identified based on established reference levels as part of the decision-making process.

The management of radioactive waste arising from the implementation of remedial measures is one component of the entire decision-making process taking into account the amounts, characteristics, properties and types of radioactive waste. The management of radioactive waste must comply with the international and national requirements for waste management facilities [39–41].

The remediation plan has to be subject to the approval of the regulatory body prior to its implementation and must state, as a minimum: the goal for the remediation; reference levels for remediation; the nature, scale and duration of the remedial measures to be implemented; the waste disposal or storage site, as appropriate; any post-remediation restrictions; and the monitoring and surveillance programmes and arrangements for institutional control for the remediation area.

The formal termination of the remediation and the release from further responsibilities of the organisation responsible for implementing the remedial measures, is based on evaluation and verification of compliance with remediation criteria. It is important to note that the termination of remediation must be subject to the formal approval of the regulatory body.

In the event that the approved remediation goals have not been met, further assessment must be performed by the responsible remediation organisation and decisions taken on whether further remedial measures or additional restrictions are required. Any modification to already approved remedial measures are subject to the approval of the regulatory body.

During the implementation of remedial measures, consideration must be given to (i) radiation safety, transport safety and waste safety, general health and safety issues and environmental issues so as to minimise hazardous

impacts, and (ii) the potential for prolonged exposure after the termination of remediation activities.

The area has to be monitored and surveyed regularly during remediation so as to verify the levels of contamination; to ensure compliance with the requirements for site release and for waste management, and to detect any unexpected levels of radiation. Where necessary, revisions to the remediation plan have to be justified and subject to the approval of the regulatory body.

Before an area can be released for unrestricted use, a survey must be performed to demonstrate that the end-point criteria and conditions, as established by the regulatory body, have been met. The organisation responsible for the surveillance and verification of activities must be clearly identified. An appropriate programme, including any necessary provisions for monitoring and surveillance, has to be established to verify the long-term effectiveness of the completed remedial measures.

If surveillance and maintenance are required after remediation is completed, a surveillance and maintenance plan must be prepared and periodically reviewed by the responsible organisation. The plan is subject to approval by the regulatory body.

A final remediation report, including any necessary final confirmation survey, must be prepared and retained by the responsible party with other records, as appropriate, and a copy submitted to the regulatory body for information.

As part of the overall management system, arrangements for archiving, retrieval and amendment of all important records concerning the initial characterisation of the area, the choice of options for remediation and the implementation of remedial measures, including all restrictions and the results of all monitoring and surveillance programmes, must be established and maintained in all cases.

3.4 Radioactive waste (RAW) management policies, regulations and standards

3.4.1 Joint Convention on the Safety of Spent Fuel Management and on the Safety of Radioactive Waste Management and other conventions

The safety of RAW is a universal concern, as illustrated by the fact that an international treaty was established in 1997 committed to achieving and maintaining a high level of safety worldwide in the management of RAW and spent nuclear fuel. This is the first and only legally binding international convention on these materials and is based on the IAEA Fundamental

Safety Principles. In 2011 the convention was supported by 58 contracting parties [49].

3.4.2 Code of Conduct on the Safety and Security of Radioactive Sources

An internationally endorsed, non-binding Code of Conduct [50] was approved in 2004 to facilitate the safe management of radioactive sources, including disused sealed sources that in most cases are declared as RAW. The objectives set out in the code should be achieved through the establishment of a comprehensive system of regulatory control of sources, applied from their initial production to their final disposal, and a system for the restoration of such control if it has been lost. To facilitate the implementation of the Code of Conduct, in 2005 a document 'Guidance on the Import and Export of Radioactive Sources of Category 1[5] and Category 2[6]' was also agreed [51]. As of May 2011, 103 Member States have expressed support for the provisions of the Code of Conduct [52].

3.4.3 IAEA safety standards

The management of radioactive waste has been under discussion at the IAEA since its establishment in 1957. In 1961 the IAEA published Safety Series No. 5, dealing with the establishment of appropriate safety procedures and practices for the disposal of radioactive waste in the sea, and in 1965 the IAEA published guidance on radioactive disposal in the ground (Safety Series No. 15). By the late 1970s, it was clear that sea disposal was not an option favoured by many countries and since then land disposal has been preferred [53]. The first formal safety standard 'Shallow Ground Disposal of Radioactive Wastes: A Guidebook' [54] was published in 1981. As mentioned earlier, the IAEA suite of safety standards is made up of a safety fundamentals publication, safety requirements standards for different activities and facilities and supporting safety guides on meeting the

[5]Category 1 sources, if not safely managed or securely protected, would be likely to cause permanent injury to a person who handled them, or were otherwise in contact with them, for more than a few minutes. It would probably be fatal to be close to this amount of unshielded material for a period of a few minutes to an hour. These sources are typically used in practices such as radiothermal generators, irradiators and radiation teletherapy [50].

[6]Category 2 sources, if not safely managed or securely protected, could cause permanent injury to a person who handled them, or were otherwise in contact with them, for a short time (minutes to hours). It could possibly be fatal to be close to this amount of unshielded radioactive material for a period of hours to days. These sources are typically used in practices such as industrial gamma radiography, high dose rate brachytherapy and medium dose rate brachytherapy [50].

requirements. The Fundamental Safety Principles publication, SF-1 [37] defines ten safety principles (see Section 3.3) that must be met for all facilities and activities involving radioactive material and ionising radiation from uranium mining through reactor operation to radioactive waste disposal. The Safety Fundamentals are supported by the following general safety requirements of relevance to spent fuel and radioactive waste management:

- Basic Safety Standards No. 115 [38], currently under revision as an interim standard No. GSR Part 3 [55] based on the ICRP103 recommendations [7];
- Governmental, Legal and Regulatory Framework for Safety, No. GSR Part 1 [56] that replaces the previous publication No. GS-R-1 [57] of 2000;
- Safety Requirements on Predisposal Management of Radioactive Waste, No. GSR Part 5 [39] that replaces Safety Requirements No. WS-R-2 on Predisposal Management, Including Decommissioning [58];
- Safety Requirements on Disposal of Radioactive Waste No. SSR-5 [40] (that combines Safety Requirements on Geological Disposal of Radioactive Waste No. WS-R-4 [59] and the Safety Requirements on Near Surface Disposal of Radioactive Waste No. WS-R-1 [60]);
- Safety Requirements on Remediation of Areas Contaminated by Past Activities and Accidents Safety Requirements No. WS-R-3 [42];
- Safety Requirements on the Management System for Facilities and Activities, No. GS-R-3 [48];
- Safety Requirements on Safety Assessment for Facilities and Activities, No. GSR Part 4 [61];
- Safety Requirements on Decommissioning of Facilities Using Radioactive Material, No. WS-R-5 [41];
- Safety Requirements on Regulations for the Safe Transport of Radioactive Material No. TS-R-1 [62];
- Safety Requirements Preparedness and Response for a Nuclear or Radiological Emergency No. GS-R-2 [63].

While the safety fundamentals and safety requirements set up provisions that must be complied with by the operators and licensees, the safety guides provide best practice for how to meet the principles and requirements. With respect to the pre-disposal and disposal of radioactive waste (i.e. GSR Part 5 and SSR-5) at present the following safety guides are of relevance:

- Classification of Radioactive Waste, No. GSG-1 [3] that substitutes the previous waste classification No. 111-G-1.1 of 1994 [64];
- Management of Low and Intermediate Level Waste, No. WS-G-2.5 [65];
- Management of High Level Waste, No. WS-G-2.6 [66];

- Storage of Radioactive Waste, No. WS-G-6.1 [67];
- Management System for the Processing, Handling and Storage of Radioactive Waste, No. GS-G-3.3 [68];
- Management System for the Disposal of Radioactive Waste, No. GS-G.3.4. [69];
- Borehole Disposal Facilities for Radioactive Waste, No. SSG-1 [70];
- Management of Waste from the Use of Radioactive Material in Medicine, Industry, Agriculture, Research and Education, No. WS-G-2.7 [71];
- Application of the Concepts of Exclusion, Exemption and Clearance, No. RS-G-1.7 [44].

In the area of decommissioning and management of waste generated during these activities, the following set of safety guides are in place:

- Decommissioning of Nuclear Power Plants and Research Reactors, No. WS-G-2.1 [45] that is currently under revision;
- Decommissioning of Medical, Industrial and Research Facilities, No. WS-G-2.2 [46] that is also currently under revision;
- Decommissioning of Nuclear Fuel Cycle Facilities, No. WS-G-2.4 [47] under revision;
- Safety Assessment for the Decommissioning of Facilities Using Radioactive Material, No. WS-G-5.2 [72];
- Release of Sites from Regulatory Control on Termination of Practices, No. WS-G-5.1 [43].

With respect to remediation and management or radioactive waste from mining and milling processing activities (past and current practices) the following IAEA safety guides apply:

- Remediation Process for Areas Affected by Past Practices and Accidents, No. WS-G-3.1 [73];
- Management of Radioactive Waste from the Mining and Milling of Ores, No. WS-G-1.2 [74] that is planned to be substituted by a new guide on Protection of the Public against Exposure to Natural Sources of Radiation including NORM (DS 421) [75];
- Occupational Radiation Protection in the Mining and Processing of Raw Materials, No. RS-G-1.6 [76].

A number of safety guides dealing mainly with spent fuel, safety assessment and safety case, as well as monitoring of disposal facilities are in a process of development and/or approval, such as:

- Storage of Spent Nuclear Fuel, No. SSG-15 [77];
- Safety Case and Safety Assessment for Predisposal Management of Radioactive Waste, DS 284 [78];
- Near Surface Disposal, DS 356 [79];

- Geological Disposal, No. SSG-14 [80];
- Monitoring and Surveillance of Radioactive Waste Disposal Facilities, DS 357 [81];
- Safety Case and Safety Assessment for Radioactive Waste Disposal, No. SSG-23 [82];
- Control of Orphan Sources and Other Radioactive Material in the Metal Recycling and Production Industries, No. SSG-17 [83].

3.4.4 EU Directives and recommendations

There are a number of European Union (EU) Directives (mandatory documents that have to be applied by member countries of the EU through their legal systems), regulations (mandatory documents that apply directly to the member countries) and recommendations (non-mandatory status). The main current directives and regulations are:

- Council Directive 96/29/Euratom on Basic Safety Standards [84], currently being revised based on the ICRP 103 [7] and the revision of the IAEA Basic Safety Standards GSR Part 3 [55];
- Council Directive 2006/117/Euratom on the Supervision and Control of Shipments of Radioactive Waste and Spent Fuel (it supersedes Directive 92/3 Euratom of 1992) [85];
- Council Regulation (Euratom) No. 1493/93 on Shipment of Radioactive Substances between Member States [86];
- Council Directive 2009/71/Euratom on Establishing a Community Framework for the Nuclear Safety of Nuclear Installations [87];
- Council Directive 2003/122/Euratom on the Control of High-activity Sealed Radioactive Sources and Orphan Sources [88];
- Council Directive 97/11/EC on the Assessment of the Effects of Certain Public and Private Projects on the Environment (amending Directive 85/337/EEC) [89];
- In addition a new Council Directive on the Management of Spent Fuel and Radioactive Waste 2011/70/EURATOM was approved in July 2011 that addresses safety at all stages of management of spent fuel and predisposal and disposal of radioactive waste [23].

3.4.5 Western European Nuclear Regulatory Authorities (WENRA) *reference levels*

The WENRA (Western European Nuclear Regulatory Authorities) reference levels are an important European initiative on the harmonisation of radioactive waste management safety, based on the IAEA safety standards and the experience of 17 countries within Europe. In the last few years the

WENRA working group on Waste and Decommissioning (WGWD) has developed safety reference levels for:

- Waste and spent fuel storage [90];
- Decommissioning [91].

The levels are based on an analysis of the current situation in countries and the different safety approaches adopted, comparison of individual national regulatory approaches with the IAEA safety standards. The initiative identified any differences and made proposals on a way to possibly eliminate the differences without impairing the resulting level of safety. The work included a review of national legal frameworks and practical implementation of the proposed WENRA reference levels. On the basis of the work performed, the WENRA member countries are working on the implementation of the agreed reference levels in their national legal framework [92].

3.4.6 National regulations

Regardless of the extensive work performed at an international level, the main responsibility for safety of spent fuel and RAW management remains with the operator/licensee and the control/oversight with the nationally designated regulatory authority. The safe development, operation and closure (siting, design, construction, operation and decommissioning or closure and subsequent control) of facilities is to a large extent dependent on an adequate regulatory framework and its effective implementation in practice. The regulatory framework will take into account the specificity of the country, interfaces between operators and regulators, and also other stakeholders.

In general three legal and regulatory approaches have been adopted, namely:

- prescriptive regulations – with very detailed and specific safety requirements (e.g., Russian Federation and the USA);
- non-prescriptive regulations – with goal oriented safety requirements (e.g., UK);
- a combination of the two above (e.g., Czech Republic, Bulgaria, etc.).

In addition specific provisions may apply in a country, e.g.:

- categorisation of spent fuel as a radioactive waste or resource;
- application of the clearance concept (e.g., that is excluded in France);
- issuance of licences, permissions or both;
- issuance of one licence for the whole life cycle of the facility or multiple licences for each stage of the facility development (siting, design, construction, commissioning, operation, closure and decommissioning/closure and institutional control).

All of the above aspects depend on the national policy and strategy adopted for radioactive waste management and nuclear development, the legal and regulatory framework within the country, past and current practices, financial mechanisms in the long term and the capacity to perform the required oversight of the facilities at present and in the future.

3.5 RAW packaging and transportation practice

International safety standards have been developed for the transport of all forms of radioactive material [62] and are issued in the form of 'transport regulations'. These 'regulations' have been adopted within all the regulations for transport of hazardous materials by all modes (land, air and sea) and are recognised and adopted in the national regulations of most countries. As with all other facilities and activities associated with RAW management and contaminated environments, the radiation safety requirements are those set down in the international Basic Safety Standards [38].

The objective of the regulations is to establish requirements that must be satisfied to ensure safety and to protect persons, property and the environment from the effects of radiation in the transport of radioactive material. This protection is achieved by requiring: (a) containment of the radioactive contents; (b) control of external radiation levels; (c) prevention of criticality; and (d) prevention of damage caused by heat. The regulations are satisfied firstly by applying a graded approach to content limitations for packages and conveyances and to performance standards applied to package designs, depending upon the hazard of the radioactive contents. Secondly, they are satisfied by imposing requirements on the design and operation of packages and on the maintenance of packagings, including consideration of the nature of the radioactive contents. Finally, they are satisfied by requiring administrative controls, including, where appropriate, approval by competent authorities. Confidence in this regard is achieved through the adoption of appropriate management systems involving quality assurance and compliance assurance programmes. The regulations are based on a classification of radioactive materials to be transported in a system of increasing hazard potential. The type of package and its testing are correspondingly higher as the hazard potential increases, with prescriptive testing and defined performance criteria for each category of package.

At the lower hazard level, low specific activity (LSA) material and surface contaminated objects (SCO) are defined quantitatively in the transport regulations. These materials can be transported in so-called 'industrial packages' (IP) of types 1, 2 and 3, which must be designed and tested according to the specifications set in the regulations. The next generic class of materials is referred to as Type A and a schedule of radionuclide specific activity

limits is provided in the regulations. Materials falling within these limitations can be transported in Type A packages for which design and testing requirements are prescribed in the regulations. The packages are designed to maintain their integrity during normal conditions of transport, providing the necessary shielding and containment, but are not expected to withstand severe transport accidents, the limitation on radioactive content ensuring that any consequences would not be severe. IP and Type A packages must conform to these design and testing requirements but do not require competent authority approval nor is notification required for international shipments of these package types. For transporting quantities of radioactive material greater than the limits for Type A packages and fissile material requires the use of Type B and C packages. These packages are designed to transport higher activity radioactive and fissile material and have to be designed with high integrity in terms of both shielding and containment features, which must be able to withstand the impacts of the most severe transport accident. Again, design and testing requirements are specified in the regulations, the latter including drop, puncture, crush and fire tests, representing the conditions that could be encountered in severe accidents, with Type C having to undergo impact testing simulating an aircraft accident in order to qualify for transporting high activity radioactive material by air. Type B and C containers require competent authority approval, both from the country of origin for Type B and also from the countries en route during the shipment.

There is no direct correlation between RAW classes and transport categories, as the classification is based on long-term safety (primarily disposal) consideration. Nevertheless, in general, low activity waste – generally VLLW from, for example, lightly contaminated building rubble from decommissioning activities – would fall in the category of LSA for transport purposes, LLW and ILW would be/could be LSA, Type A or Type B and HLW would be transported as Type B material.

3.6 Conclusion

Significant progress can be observed in the development of internationally agreed standards on the management of RAW, radiation safety and transport safety in recent years. The role of each country is to implement these standards in the most efficient and appropriate manner, taking into account the specific characteristics and conditions of existing RAW or anticipated future arisings. One of the main challenges is for operators and regulators to apply a graded approach based on the existing and potential risks to the public and the environment and at the same time providing confidence in the demonstration of adequate levels of safety.

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4

Technical solutions for the management of radioactive waste (RAW): overview and methods of selection

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Abstract: Technical options for waste streams which arise from nuclear applications, research, power generation, nuclear fuel cycle activities and decommissioning of nuclear facilities as well as NORM-containing waste, are summarized. Since optimal selection of technical options is case specific to the waste management needs, they are not ranked. However, selection criteria for waste processing and disposal technologies are summarized and a systematic approach for selection of optimal solutions is proposed.

Key words: waste management, processing, disposal, classification, categorization, waste routing.

4.1 Introduction

Waste management is a subject that has received considerable attention and is recognized as an important link for public acceptance of nuclear energy and its applications. Technical options and technologies are crucial for safe management of radioactive waste. A wealth of information is currently available about a multitude of waste management technologies and their technically novel and alternative designs, as well as about emerging technologies, which require further development and/or validation. Selection among available options and technologies can be done on a national level, or by waste generators or by waste management organizations. The selection principles may vary by organizational preference, collected or known experience or following an optimization procedure. In any case, because of the costs involved, the potential complexity of technical and environmental considerations, as well as the necessity to ensure adequate performance, the selection mechanism will always require rather clear criteria in order to address waste management needs. Some criteria will be fairly general and applicable to almost any waste management system. Others may apply to specific waste categories or to particular waste management steps.

The aim of this chapter is to summarize technical options for waste streams which arise from nuclear applications, research, power generation, nuclear fuel cycle activities and decommissioning of nuclear facilities as well as naturally occurring radioactive materials (NORM)-containing waste and to propose a systematic approach for selection of optimal solutions. IAEA publications [1–3] form the basis for establishing appropriate strategies and infrastructure for the management of radioactive waste. The infrastructure requires selection of an optimized technology/option because of the variety of processes and techniques available for different waste streams at specific waste management steps. The technologies selected for different waste management steps should then be combined in an integrated strategy to optimize the overall waste management system [4]. The selection of waste technologies for each specific waste stream/category should be based on an evaluation process with the following elements:

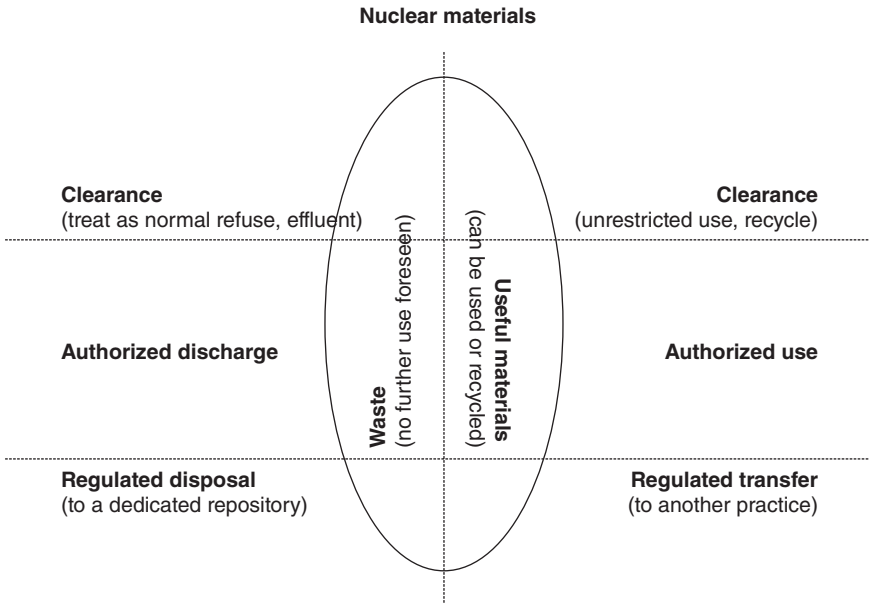
- identification and nature of specific radioactive waste inventories and associated properties;
- consideration and review of various options for the management of that waste;
- evaluation of the advantages and disadvantages of each option using multi-attribute utility analysis (MUA) [5] or any other suitable methodology that compares safety, technological status, cost-effectiveness and social and environmental factors;
- selection of the best available technology(ies) not entailing excessive cost and satisfying all regulatory requirements [6];
- approval (via licensing, authorization) of the selected technology(ies).

4.2 Waste routing, classification and categorization

The main nuclear material routes are (Fig. 4.1):

- clearance from regulatory control, which assumes unrestricted disposal of waste and unrestricted reuse of useful materials;
- authorized release, which assumes authorized discharge of waste to the environment and authorized reuse of useful materials;
- regulated disposal of waste and regulated transfer of useful materials to other practices.

There is great diversity in the types and amounts of radioactive waste in different countries. Technologies for management of the waste are also diverse, although the main technological approaches are likely to be similar everywhere. Adequate processes and technologies can be identified based on detailed information about the current or forecast waste, e.g. waste classification, categorization, properties and inventory.



4.1 Schematic of radioactive materials routing (radioactive waste road map).

The IAEA provided an internationally accepted waste classification system [7] which define the following classes according to the activity and half-lives of radionuclides in waste:

- exempt waste (EW);
- very short lived waste (VSLW);
- very low level waste (VLLW);
- low level waste (LLW);
- intermediate level waste (ILW);
- high level waste (HLW).

The IAEA classification is based primarily on long-term safety and therefore is oriented on the selection of the most appropriate disposal routes (end-points) for solid or solidified waste.

Management of waste in different steps prior to disposal (e.g., pretreatment, processing, storage) requires complementary information on the waste properties relevant to particular activities. Categorization of waste is used to provide a consistent approach to waste processing and storage. Categorization of waste has to include information such as origin, physical state, types, properties and process options [8, 9].

4.2.1 Waste inventory

Several levels of detail of waste inventory may be required. For example, a waste processor would need a more detailed inventory which will suggest management methods for each and every particular waste type. For national waste management agencies, less detail is needed to formulate future plans. A more generic inventory is required for policy makers and developers of a national waste management strategy. Ref. [2] provides practical guidance and suggests methodologies for preparing national and waste owner inventories of radioactive waste.

4.3 Waste management steps

Waste management is typically divided into pre-disposal and disposal steps. Pre-disposal comprises all the steps in the management of radioactive waste (RAW) from its generation up to disposal, including processing (e.g., pre-treatment, treatment and conditioning), temporary (interim) storage and transport. Disposal envisages permanent emplacement of waste in an appropriate facility without the intention of retrieval. Radioactive waste is prepared for disposal by processing technologies primarily intended to produce a waste form that is compatible with the selected or anticipated disposal option. For evaluation of a particular process or technology, it is necessary to review the availability of selected options to meet waste processing, storage and disposal requirements.

The life cycle of radioactive waste consists of a number of steps:

- *Pre-treatment* includes any operations prior to waste treatment, to allow selection of technologies that will be further used in processing of waste (treatment and conditioning), such as: collection, segregation, decontamination, chemical adjustment and fragmentation [10].
- *Treatment* of RAW includes those operations intended to improve safety or economy by changing its characteristics. The basic objectives of treatment are: volume reduction; radionuclide removal from waste; and change of physical and chemical composition. Treatment includes operations intended to benefit safety and/or economy by changing the characteristics of the waste [11]. Some treatment may result in an appropriate waste form. However, in most cases the treated waste requires further conditioning either by solidification, immobilization or encapsulation.
- *Conditioning* covers those operations that produce a waste package suitable for handling, transportation, storage and/or disposal. It may include: immobilization of the waste; enclosure of the waste in containers; and, if necessary, provision of an overpack. Immobilization refers to

the conversion of waste into a waste form by solidification, embedding or encapsulation. Common immobilization matrices include cement, bitumen and glass.

- *Storage* of RAW involves maintaining it such that retrievability is ensured and confinement, isolation, environmental protection and monitoring are provided during the storage period.
- *Transportation* refers to the deliberate physical movement of RAW in specially designed packages from one place to another. For example, raw waste may be transported from its collection point to centralized storage or a processing facility. Conditioned waste packages may be transported from processing or storage facilities to disposal facilities.
- *Disposal* envisages emplacement of waste in an appropriate facility without the intention of retrieval. Note that in some countries controlled discharge of effluents to the environment is often considered as a regulated disposal option.
- *Characterization* of RAW is an important aspect at every stage of waste management. It involves determination of the physical, chemical and radiological properties of the waste to establish the need for further adjustment, treatment, conditioning, or its suitability for further handling, processing, storage and disposal. Up-front characterization as part of the pre-treatment stage is essential for technical decision making involving the selection of the most efficient treatment process. Methods of RAW characterization and the methodology of characterization, including sampling procedures, are described in detail in Refs [8] and [9].

4.4 Technical options for waste management

4.4.1 Gaseous and airborne waste treatment

Operations involving the handling of radioactive material may generate airborne radioactive contamination. The basic difference between airborne effluents and RAW in condensed (i.e., liquid or solid) phase is that airborne material has no definite volume and its dispersion in the environment is very fast. Gaseous and airborne wastes are discharged to the environment through ventilation and air-cleaning systems, which are a vital part of the general design of a nuclear facility [11–14]. The combination of a well-designed ventilation system with thorough cleaning of exhaust air is the main method of preventing radioactive contamination of the air in working areas and in the surrounding atmosphere. Ventilation and air-cleaning systems should provide efficient treatment of gaseous streams under normal operations, maintenance and accident conditions. High efficiency particulate air (HEPA) filters are most commonly used for removal of radioactive

particulates and aerosols from gaseous streams [12, 14]. Sorption beds charged with activated charcoal are common for removal of volatiles (e.g. iodine) and as delay beds for noble gases. Wet scrubbers are used for the removal of gaseous chemicals, particulates and aerosols from process off-gases. Additional components of the air-cleaning system include pre-filters, and temperature and humidity control systems, as well as monitoring equipment such as gauges that show pressure differentials. The treatment of gaseous streams results in secondary waste either solid (spent filters or sorption beds) or liquid (scrubbing solutions). The physical and chemical properties of the selected air-cleaning media should therefore be compatible with the treatment and conditioning processes for the solid or liquid waste streams in which they will be treated.

4.4.2 Aqueous waste treatment

In most cases treatment of aqueous waste aims at splitting it into two streams: (a) a small fraction of concentrate containing the bulk of radionuclides; and (b) a large part, the level of contamination of which is sufficiently low to permit its discharge to the environment or for recycling [11]. Effective liquid treatment separates as much of the radioactive contamination as possible from the waste in a concentrated form. Generally, the radioactive concentrate requires additional conditioning prior to disposal. Aqueous treatment processes are usually based on conventional physical and chemical treatment principles with individual characteristics of the waste to be considered. Failure to account for the chemical and biological nature of aqueous waste may result in inadequate treatment and/or conditioning and could even damage the waste processing facilities. Detailed descriptions of the technologies can be found in Refs [11, 15–18]. Historically, three technologies have mainly been applied to treat aqueous waste, namely *chemical precipitation*, *ion exchange* and *evaporation*. *Membrane processes* such as reverse osmosis, nanofiltration, ultrafiltration and microfiltration are now also successfully used and demonstrating good performance. In each case, process limitations due to corrosion, scaling, foaming and the risk of fire or explosion in the presence of organic material should be carefully considered, especially with regard to the safety implications of operations and maintenance. If the waste contains fissile material, the potential for criticality should be evaluated and eliminated to the extent practicable by means of design and administrative features.

The objective of a *chemical precipitation* process [15] is to remove radionuclides from liquid waste by the use of an insoluble finely divided solid material. The insoluble material, flocculate or floc is generally, but not necessarily, formed *in situ* in the waste stream as a result of a chemical reaction. The use of these processes concentrates the radioactivity present in a liquid

waste stream into a small volume of wet solids (sludge) that can be separated by physical methods from the bulk liquid component. Chemical precipitation is suitable for the waste which is low in radioactivity, alkaline in pH and contains a significant salt load. This process is simple and relatively inexpensive in terms of the plant and its operation but it requires good understanding of the process chemistry and strict consideration of process parameters. The process may be limited by the activity level.

Ion exchange is a standard method of liquid clean-up [16]. The ion exchange materials are insoluble matrices containing displaceable ions, which are capable of exchanging with ions in the liquid passing through by reversible reaction. Organic and inorganic, naturally occurring and synthetic ion exchangers have found their specific fields of application in different purification and liquid waste treatment processes. If the waste is relatively free of salts, mildly acidic in pH and requires a decontamination factor of around 100 or so, ion exchange may be a good choice. This process is more expensive than chemical treatment – especially when special purpose resins are used – but has a wider range of application with regard to radioactivity concentration.

The limitation of conventional filtration and ion exchange is that colloidal particles, some radioactive, pass straight through to the product (treated) water. Colloidal particles containing $^{58/60}$Co, ^{54}Mg, ^{55}Fe and ^{125}Sb are typical examples. Ultrafiltration is capable of removing these particles completely and has been adopted at a number of sites to complement the existing conventional filtration/ion exchange systems.

Membrane processes [17] are successfully used as one or more of the treatment steps in complex waste treatment schemes, which combine conventional and membrane treatment technologies. For example, *electrodialysis* is a well-established membrane technology that has been used widely for the desalination of brackish water and also to separate monovalent ions from multivalent ions. These combined systems offer superior treatment capabilities, particularly in instances where conventional methods alone could not perform a similar task as efficiently or effectively. They are capable of producing high-quality treated effluents with an acceptably low level of residual radioactivity for discharge, or for recycle and reuse. The concentrate waste stream containing the removed radioactivity invariably needs further processing by evaporation or other means to facilitate final conditioning to a solid waste form suitable for intermediate storage and disposal. When applying membrane technologies, the selection of the membrane material, its configuration and the operating parameters are critical. A wide variety of membranes are commercially available with different operational characteristics [17]. The choice of a membrane must be based not only on performance data (salt rejection, flux), but also take into account the interaction of the membrane with the feed solution and whether this will lead

to stable operation and minimal fouling (a process where deposits on surface or into pores of membrane cause performance degradation).

Evaporation is a proven method for the treatment of liquid radioactive waste providing both good decontamination and good concentration [18]. Water is removed in the vapour phase of the process leaving behind non-volatile components such as salts and most radionuclides. There could be situations when waste volumes are somewhat high, having a low salt content but a considerably higher activity level; in this event evaporation is used to reduce the waste volume to a concentrate and also to obtain a high decontamination factor (of the order of a few thousand). However, the process can be limited by the presence of volatile radionuclides, and also it is energy-intensive.

4.4.3 Organic liquid waste treatment

The organic nature of the waste often introduces additional hazards not encountered with inorganic waste, such as susceptibility to radiolysis and biodegradation, flammability, volatility, chemical toxicity and inherent biohazards. This results in special requirements and considerations for storage, treatment, conditioning and disposal of this waste. The volume of organic liquid waste is usually small compared with aqueous RAW. Unlike aqueous waste, it may not be possible to discharge treated organic waste to the environment because of its organic chemical content. The goals of organic liquid waste treatment may be: (a) conversion to a solid form either directly or after chemical adjustment to a form compatible with a solidification matrix (e.g., cement); (b) volume reduction; (c) decontamination for reuse. Various techniques for the treatment of liquid organic waste have been developed and implemented in different countries [19, 20]. In some cases processes and equipment selected for the treatment of aqueous and solid waste had been adapted for processing organic liquid waste, and combined processing was cost effective. For example, small quantities of organic liquid can be readily mixed with solid waste in an incinerator. Properly controlled *incineration* is an attractive technique for treating organic liquids because they are readily combustible, and high volume reduction factors can be achieved. After combustion, radionuclides from the waste will be distributed between the ash, filters and off-gas, to a degree which depends on details of the unit's design and operating parameters. Further immobilization, such as grouting of ashes, will be required to stabilize these residues, some of which will have a much higher radionuclide concentrations per unit volume compared to the original waste.

Wet oxidation is a technique for breaking down organic materials to carbon dioxide and water in a process that requires significantly lower temperatures compared to incineration. The main advantages of the process

are the low temperatures required and use of aqueous media, which is easy to treat.

Simple *distillation* may be used for the treatment of scintillation fluids and miscellaneous solvent waste. Substantial volume reduction is possible and the recovered organic solvent could be used as a technical grade solvent or as a fuel for an incinerator. Distillation can be practised with conventional readily available equipment and space requirements for the equipment are small.

There are difficulties with solidification of organic waste by cementation. Only about 12 vol.% of oil can be incorporated directly in cement and still retain a waste form that is dry and monolithic. However, significant increases in waste loadings can be obtained when *emulsification* is applied. A simple way of on-site treatment of organic liquid RAW is *converting the liquid to a solid form with absorbents*. As long as there is an excess of absorbent, there is no need even for mixing; the liquid waste can be added to the absorbent in a suitable container and eventually all the liquid will be taken up. This technique has been routinely used for the solidification of radioactive turbine and pump oil. The use of absorbents converts the liquid waste into a form, which can vary from loose dry particles to a jelly-like solid. The waste forms have no special integrity and are only restrained from dispersing by the container. Another frequently used option is embedding of absorbed organics into a cementitious waste form.

4.4.4 Solid waste treatment

The essential purpose of solid low and intermediate level waste treatment is to reduce the volume. Both compaction and supercompaction technologies are described in detail in Ref. [21]. *Compaction* involves compressing the waste into containers or boxes in order to reduce the volume. Low force compaction is the least expensive and an easier volume reduction process than high force compaction. Compaction units are also amenable to automation, which can improve operational efficiency and radiation protection aspects. High force compactors can give somewhat better reduction factors, whereas supercompactors achieve highest volume reduction which is close to the theoretical density of materials by minimizing the voidance. Both high force and supercompactors typically compress the waste inside of drums. From a technical viewpoint, the same technique may be applied as a treatment or a pre-treatment step, depending on the required sequence in the overall waste management scheme. For instance, shredding could be considered as treatment when applied for volume reduction of waste before packaging, and as pre-treatment when applied before incineration. In another example, low pressure compaction may be applied as a treatment method when used before supercompaction as a pre-compaction step.

Thermal treatment (incineration, pyrolysis, plasma, etc. [11, 21, 22] may provide the best potential for effective volume reduction of generated solid waste. A further advantage of employing thermal treatment is an improvement of homogeneity and quality of the waste form obtained after treatment and conditioning. Considering the high overall costs of waste disposal and the growing requirements for improved quality of the final waste form, the benefits offered by thermal processing become very significant. Thermal methods may also have disadvantages restricting their applications. The primary consideration is meeting environmental safety requirements, such as gaseous effluent restrictions, which lead to greater complexity and higher costs of these technologies. Higher implementation costs may not justify the application of incineration for relatively small volumes of solid waste. Generally, the permits or licences from regulatory agencies will stipulate numerical emission limits or known reference standards to be met. *Melting* waste metal scrap, with resultant homogenization of the radioactive material and its accumulation in the slag, may be considered as a means of achieving authorized reuse or removal of regulatory control [22].

Two *emerging technologies*, molten salt oxidation and thermochemical treatment, have demonstrated promising performance parameters, although to date they have limited application. Molten salt oxidation is a flameless thermal desorption process [22]. The waste is introduced into a bath of molten salts, typically at temperatures between 500 and 950°C. This has the effect of oxidizing the organic constituents of the waste. Carbon dioxide, nitrogen and water are produced. The end product is an organic-free salt residue which captures radionuclides, metals and other inorganics. The production of acid gas emissions is inhibited by the formation of the stable salts. Thermochemical treatment uses powdered metallic fuel (PMF), such as Al or Mg, which interacts with the waste both chemically and physically through reaction with the water present in the waste. This results in the formation of hydrogen gas and heat; the subsequent combustion is used to destroy the organic material, resulting in solid slag or ash. The hydrogen gas burns because of the presence of enough oxygen, and in co-reaction the waste is combusted leaving a slag-like product. The presence of excess metal powder suppresses the production of corrosive gases. Thermochemical processing technologies are used for treating and conditioning problematic radioactive wastes [23]. Thermochemical processing uses PMFs that are specifically formulated for the waste composition and react chemically with the waste components. The composition of the PMF is designed to minimize the release of hazardous components and radionuclides in the off-gas and to confine the contaminants in the ash residue. Thermochemical procedures allow decomposition of organic matter and simultaneous capture of hazardous radionuclides and chemical species [22, 23].

4.4.5 Bio-hazardous waste treatment

Some solid and liquid wastes may contain bio-hazardous or infectious materials. Further to radiological protection, other precautions for handling these wastes should be respected. When processing bio-hazardous wastes, their infectious features, and tendency to putrefaction, to insect attacks and to microbial degradation must be controlled. Clearance of bio-hazardous waste from radiation regulatory control is unlikely to mean that this waste is also exempt from bio-hazardous waste regulatory control. The goals of treatment of bio-hazardous waste are the following: (a) biologic detoxification; (b) prevention of biological degradation; (c) volume reduction.

An important step in the treatment of bio-hazardous waste is neutralization of biological hazard. It can be done by sterilization. A number of sterilization methods are regularly used in hospitals and they can be applied for treating bio-hazardous RAW with some adaptation. Some other methods are aimed at volume reduction of the waste. Available treatment methods for bio-hazardous and medical radioactive waste have been described in detail [24]. Lidded containers lined with plastic bags are used for collection of wastes displaying biological hazards. Special consideration should be given to sharp objects. When possible, these items should be collected in puncture-resistant packages, properly labelled and treated separately. Most microbiologically contaminated laboratory wastes are suitable for *steam autoclaving*, but this method should not be used where the radioactive content of the waste is volatile during steam treatment. This method is not considered appropriate for most non-microbial pathogens, animal carcasses or parts. *Chemical disinfection* is useful for laboratory ware or similar materials, but it is not suitable for pathological waste and animal carcasses or parts. *Gamma irradiation* is an attractive option for sterilization since it is appropriate for pathological waste, animal carcasses and parts. After deactivation or procedures aimed at preventing decomposition of its biological components, bio-hazardous waste can usually be treated using the same methods applied to non-biological radioactive materials in order to meet the waste acceptance criteria. *Incineration* is the preferred method for treating bio-hazardous RAW of animal or human origin, as well as organic chemical waste [11, 21]. Incineration provides complete combustion of waste, producing totally sterile residues, with any emissions from the stack being kept to acceptable environmental standards. *Thermochemical treatment* has been proven to be an effective method to treat animal carcasses producing totally sterile slag residue, with minimal off-gas emissions, the composition of which can be kept in line with acceptable environmental standards [11, 22, 23].

In cases where incineration is not available or the volumes of human and animal wastes are so low that it is desirable to treat them as they are produced, it may be feasible to use *maceration/pulverization* to render these

materials liquid, so that they can be discharged via a liquid RAW route, including any necessary chemical deactivation to treat the biological hazard. Compaction and shredding are not considered viable for treatment of biohazardous solid waste. The primary reason for this restriction is that any microorganisms contained within the waste may be spilled or released during these processes and contamination may be widely dispersed.

4.4.6 Spent fuel treatment

Spent nuclear fuel (SNF) may be considered either as waste (SFW), which will eventually be packaged and disposed of [25], or reprocessed to recover uranium and plutonium followed by conditioning of residue in the form of high level waste (HLW) containing mainly fission and activation products, and so-called minor actinides (Np, Am, Cm) [11, 25]. There is no specific treatment step of SFW except that fuel elements are stored at the reactor site for some time to allow their intense radioactivity to decay and associated heat to decrease. SFW elements can then be moved to longer term storage facilities (dry or wet), before deep geological disposal. If not declared as waste, SNF elements could be shipped to a reprocessing plant, but only after a suitable storage (cooling) period. The decay/cooling storage period at reactor sites usually varies from three to five years or even longer; afterwards, the spent fuel can be transferred to 'away from reactor' storage for up to 50 years or more, depending on the national policies with regard to reprocessing or disposal.

HLW formed after reprocessing of SNF contains fewer long-lived actinides than SNF due to extraction of plutonium. The world industrial reprocessing practice (as used, for example, at the La Hague Reprocessing Plant in France) demonstrates that the volume of HLW after conditioning is less than the total volume of SNF assemblies [26]. Both LLW and ILW generated by reprocessing are treated by methods described in this chapter. HLW contains some long-lived fission products such as Tc-99 and I-129, and minor actinides (Np, Am, Cm). An additional technological procedure – partitioning – can be introduced into reprocessing technology for extraction of minor actinides to reduce the HLW radiotoxicity. The extracted minor actinides could then be transmuted by fission using fast neutrons. The partitioning and transmutation (P&T) approach can reduce the radiotoxicity of SNF by a factor of 100 or more [27] resulting in less dangerous waste for disposal.

4.4.7 NORM waste treatment

Conventional industries generally produce large volumes of residues containing naturally occurring radioactive materials (NORM), of the order of

10^4–10^6 t/a. This necessitates a different, pragmatic approach from typical RAW management, for which the principle of concentration and containment is used [28]. For most NORM residues containment is not possible, and in many cases it is not waste but a useful recyclable residue. Therefore, for NORM residues the principle of dilution and dispersion should be preferred wherever possible. It saves resources of other materials and it reduces waste volumes. Furthermore, one should keep in mind that concentration/containment and dilution/dispersion are complementary, not contradictory, concepts. Processing of NORM waste consists of pile stabilization by various processes in order to increase the safety of storage and disposal sites. Large solid pieces of NORM waste, such as pipes from the oil industry, are fragmented for handling and transport purposes. Liquid effluents are generated at all stages of the uranium production cycle that use process water and chemicals, including crushing, grinding, leaching, precipitation and tailings disposal and management. In addition, leaching of ore and mineralized waste rock by groundwater and surface water, respectively, can result in generation of acid mine water, which must also be contained and treated. The effluents contain radioactive and non-radioactive elements and compounds that, if not properly contained, can contaminate drinking water resources or enter the food chain, potentially harming the environment and endangering the health and well-being of human populations. Scales and sludges, which are generated in small volumes but which may have activity concentrations reaching very high levels, such as those originating from the oil and gas industry, are usually held in storage pending the establishment of suitable disposal facilities [28].

Criteria for exemption, without further consideration, of substances containing radionuclides of artificial origin are based on the premise that exemption will be the optimum option when the dose incurred by an individual is of the order of 10 μSv or less in a year [11]. For NORM, the situation is quite different. Owing to the existence of significant and highly variable levels of background exposure to radionuclides of natural origin, exemption is likely to be the optimum option over a much wider range of doses, typically doses of the order of 1 mSv or less in a year.

The use, reuse and recycling of NORM residues and NORM contaminated items – including, where appropriate, the dilution of NORM residues to reduce the activity concentration – is now starting to be recognized as a legitimate and desirable option for minimizing the quantities of NORM that need to be disposed of as waste. In particular, the beneficial (and safe) uses of phosphogypsum as a co-product of fertilizer production are now very much in the spotlight and, in some countries at least, there is already evidence of a shift in regulatory attitude towards this approach. However, when considering the use of NORM residues in the construction of dwellings, as a component of either landfill material or construction material, the

possibility of increased radon exposure needs to be carefully taken into account.

4.4.8 Conditioning: waste forms

Conditioning includes those operations that produce a waste package suitable for handling, transport, storage and/or disposal [11]. Conditioning may include the conversion of the waste to a solid waste form, additional immobilization of some solid waste, packaging of the waste form into containers, and, if necessary, providing an overpack. The *waste form* is the waste in its physical and chemical form after treatment and/or immobilization (resulting in a solid product) prior to packaging. The waste form is a component of the waste package. The *immobilization* of RAW (solidification, embedding or encapsulation) to obtain a stable waste form is an important step in waste management needed to minimize the potential for migration or dispersion of radionuclides into the environment during storage, handling, transport and disposal. Radioactive and chemically hazardous constituents in the waste can be immobilized into a waste form material through two processes: Constituents can be (1) bound into the material at atomic scale (*chemical incorporation*), or (2) physically surrounded and isolated by the material (*encapsulation*) [11].

A number of matrices have been used for waste immobilization and those include glass, ceramic, cement, polymer and bitumen [11, 12, 21, 29–36]. The choice of the immobilization matrix depends on the physical and chemical nature of the waste and the acceptance criteria for the storage and disposal facilities to which the waste will be consigned. Several factors must be considered when selecting a waste form material for immobilizing a specific waste stream. The key considerations include the following [11, 21, 33, 34]:

- *Waste loading*: The waste form must be able to accommodate a significant amount of waste (typically 25–45 w%) to minimize volume, thereby minimizing the space needed for storage, transportation and disposal.
- *Ease of production*: Fabrication of the waste form should be accomplished under reasonable conditions, including low temperatures and, ideally, in an air atmosphere, using well-established methods to minimize worker dose and the capital cost of plant.
- *Durability*: The waste form should be physically durable and have a low rate of dissolution when in contact with water to minimize the release of radioactive and chemical constituents.
- *Radiation stability*: The waste form should have a high tolerance to radiation effects from the decay of radioactive constituents. Depending on the types of constituents being immobilized, the waste form could be subjected to a range of radiation effects, including ballistic effects from

alpha decay and ionizing effects from decay of fission product elements.

- *Chemical flexibility*: The waste form should be able to accommodate a mixture of radioactive and chemical constituents with minimum formation of secondary phases that can compromise its durability.
- *Availability of natural analogues*: Since direct laboratory testing of the waste forms over the relevant timescales for disposal (typically 10^3–10^6 years) is not possible, the availability of natural mineral or glass analogues may provide important clues about the long-term performance of the material in the natural environment, thereby building confidence in the extrapolated behaviour of the waste form after disposal.
- *Compatibility with the intended disposal environment*: The waste form should be compatible with the near-field environment of the disposal facility. The near-field environment provides the physical and chemical conditions that are favourable for maintaining waste form integrity over extended periods, which helps to slow the release of constituents and their transport out of the facility.

A number of materials have been used for waste immobilization and those include glass, ceramic, metal, cement, polymer and bitumen. All these materials have their advantages and disadvantages both in terms of the kinds of waste that can be immobilized and the properties of the solidified waste forms obtained. The choice of the immobilization matrix depends on the physical and chemical nature of the waste and the acceptance criteria for the disposal facility to which the waste will be consigned.

Glass is being used worldwide to immobilize HLW from reprocessing of spent nuclear fuel and targets [11, 21, 35]. The immobilization process, *vitrification*, is a continuous process capable of handling large-volume waste streams. Vitrification has demonstrated its efficiency and its flexibility in a number of countries. It has become the reference process for the conditioning of HLW and is currently deployed on a large scale for lower activity waste streams. Given the good results obtained with vitrification of HLW, several projects are underway for the vitrification of slurries, low and intermediate level solid waste, mixed waste, etc. The advantages are: volume reduction, destruction of organic constituents including hazardous materials, immobilization of radioactive and hazardous components, advantages for storage, transportation and disposal. The vitrification processes are sufficiently robust, which means that they accept almost any waste after a minimum of up-front characterization, with reproducible characteristics of the end product and acceptable off-gasses. Vitrification can also be performed *in situ* as a special case (e.g., legacy waste or contaminated soil) [23].

Crystalline ceramics are inorganic, non-metallic solids that contain one or more crystalline phases. Single-phase crystalline ceramics can be used to

immobilize separated radionuclides (e.g., plutonium-239) or more chemically complex waste streams (e.g., HLW) [11, 36]. In the latter case, the atomic structure of the ceramic phase must have multiple cation and anion sites that can accommodate the variety of radionuclides present in the waste stream. These materials are potentially attractive for immobilizing long-lived alpha-emitting actinides such as plutonium, neptunium and americium. However, some of these materials are susceptible to radiation damage effects associated with alpha decay from actinides [36]. Multiphase crystalline ceramics (e.g., Synroc) consist of an assemblage of crystalline phases. Individual phases are selected for the incorporation of specific radionuclides, with the proportions of phases varying depending on the composition of the waste stream. An individual phase can host one or more radionuclides, including solid solutions of radionuclides. However, not all phases will host radionuclides. Ceramic materials and methods of fixation are largely at an early stage of developement. Ceramic products are crystalline in nature and therefore thermodynamically stable although they are sensitive to radiation damage.

Glass-composite materials (GCMs) are materials that contain both crystalline and glass phases [11, 21, 34, 35]. Depending on the intended application, the major component may be a crystalline phase with a vitreous phase acting as a bonding agent. Alternatively, the vitreous phase may be the major component with particles of a crystalline phase dispersed in the vitreous matrix. GCMs can be formed by a number of processes, including melt crystallization (controlled or uncontrolled), multiple heat treatments, or by encapsulation of ceramic material in glass. GCMs offer several potential advantages over glass for use as waste form materials, including increased waste loadings, increased waste form density, and thus smaller disposal volumes. These waste forms can also be used to immobilize glass-immiscible components such as sulphates, chlorides, molybdates, and refractory materials that have very high melting temperatures. They can also be used to immobilize long-lived radionuclides (e.g., actinides) by incorporating them into the more durable crystalline phases; short-lived radionuclides (e.g., many fission products) can be accommodated in the less durable vitreous phase [35].

Cements are inorganic materials that set and harden as a result of hydration reactions [11, 21]. Cements are used to immobilize waste having relatively low levels of radioactivity (i.e., low or intermediate level RAW). Higher activity wastes can result in radiolysis and production of hydrogen gas from the breakdown of water or hydroxyl groups in the cement. Cementation is viewed as low cost and forms a major part of both solid and liquid (mainly aqueous) LLW and ILW immobilization technologies. The range of applicability of cements is to be considered in view of the characteristics of the environment and of the initial waste. The cement may

display pH buffering properties and, consequently, control mobility of most radionuclides in the disposal environment. The quality of cemented waste forms is continually being improved. They are efficient for the immobilization of alpha-bearing waste. The problematic side is the potential for reaction with some wastes and the relatively high porosity and leachability for some radionuclides of the end-product. In addition, cementation results in a volume increase rather than decrease, so a smaller than unit reduction factor.

Bitumen, a viscous hydrocarbon and a major component of asphalt, has been used to solidify and stabilize radioactive materials [11, 32]. Bitumen immobilizes waste mainly by encapsulation rather than binding the waste chemically. The advantages of bitumen as a waste form are simplicity of production, low operating cost, and leach-resistant characteristics. However, bitumen can be a fire hazard, especially when oxidizing wastes like nitrates are involved.

Some waste treatment methods, such as plasma arc melting, or molten metal techniques, result in both a high volume reduction and very stable waste forms.

When spent fuel is not reprocessed (the once-through fuel cycle), SFW conditioning consists of volume optimization (rearrangement of the fuel rods) and enveloping them in a multi-component barrier consisting of various *metals* (copper, lead) and the packaging canister [25]. Several different types of metallic materials have been studied as potential waste forms. Like crystalline ceramics, metal waste forms can consist of single- or multiple-phase assemblages and the waste form itself can be granular or monolithic. Metal waste forms can be fabricated by sintering or casting. Each of these techniques has drawbacks; in particular, it can be difficult to find metal compositions and processes that effectively wet and encapsulate dispersed phases or fines.

4.4.9 Conditioning: waste packages

The *waste package* is the product that includes the waste form and any container(s) and internal barriers (e.g., absorbing materials and liner), as prepared in accordance with requirements for handling, transport, storage and/or disposal [11]. These requirements can be different for each step indicated above or they can be combined in one set of parameters that combine conservative requirements for each step. If there may be a significant delay before an acceptable disposal route becomes available, the container should provide integrity during the pre-disposal storage period and should be capable of allowing for: (a) retrieval at the end of the storage period; (b) transport to and handling at a disposal facility; (c) performance as required in the disposal environment.

If a container is not initially designed to meet the relevant acceptance criteria for transport, storage or disposal, an additional container, or an overpack, will be necessary to meet the acceptance criteria. Care should be taken to consider the compatibility of the waste package and the overpack with respect to the waste acceptance criteria (WAC). Waste packages are often produced when no disposal facility exists and therefore no applicable disposal WAC are available to guide the design and preparation of the packages [11, 21]. In this case, it may be necessary to develop waste package specifications in place of the WAC. These specifications are considered as a design output, and are intended to control the radiological, physical and chemical characteristics of the waste package to be produced. Waste specifications are usually oriented towards the performance or control of specific facility processes and may be used as a contractual vehicle to control subcontracted operations. Waste specifications, like the WAC, should be cognizant of intended storage/disposal facility parameters and transport regulations, and incorporate relevant parameters of the WAC, or in lieu of the WAC, when they have not been developed. It should be noted that the requirements on waste packages imposed by the IAEA transport regulations [37] meet many of basic requirements of the generic WAC.

4.4.10 Waste storage

Storage should be provided for conditioned waste as well as for untreated/unconditioned (raw) waste. The matrix in which the RAW are immobilized will have a significant effect on the properties of the waste package and can strongly influence its required performance [21]. RAW to be placed for storage has to comply with certain requirements established by the operator of a storage facility. In general, these requirements can be formulated in the following way:

(a) the waste should be packaged in such a way that the package integrity can be assured during the entire planned storage time;
(b) the surface dose rate and contamination level of the storage package should comply with the requirements of the storage facility;
(c) each storage package shall be uniquely and durably identified.

The design features of storage facilities may vary greatly depending on the objective of storage, characteristics of the stored waste and period of storage [38]. The design of storage facilities has to meet national regulatory standards and basic safety principles. In this context, the facility should be capable of maintaining the 'as-received' integrity of the waste package until it is retrieved for disposal. The storage facility is protecting the waste from environmental conditions, including extremes of humidity, heat and cold, or any other environmental condition, which would degrade the waste form or container. Local climatic conditions may result in the need for cooling

or dehumidifying of the store atmosphere, in order to avoid possible deterioration of the waste packages.

As far as the siting of a storage facility is concerned, it should be situated above the groundwater level, and certainly not in a flood plain. In areas of high rainfall, the facility should be constructed with appropriate systems to protect against intrusion of groundwater. Waste storage facilities by design vary from a simple steel safe to a sophisticated engineered facility. Storage of untreated waste materials is currently practised, mainly for practical reasons (e.g., to allow short-lived radionuclides to decay) or as a buffer in view of optimal use of the treatment facilities. Storage for decay is particularly important for RAW resulting from medical uses of radioisotopes since many radioisotopes are short-lived and the activity of the waste produced is well defined. Practical experience shows that on-site decay storage is suitable for waste contaminated with radionuclides with a half-life up to 100 days. Where large volumes of short-lived radioactive wastes are produced, it may be more convenient to partition the short-term decay storage facility to provide areas for storage of wastes according to their half-life. Storage of conditioned RAW in engineered facilities is characterized by the fact that it is controlled, that the material is retrievable, that maintenance and, if required, secondary packaging (overpack) remain possible and, eventually, that the material can be transferred to a final location to be decided in due time. Conditioned waste including spent nuclear fuel, vitrified HLW and long-lived waste has been safely and securely stored in a number of countries for several decades. Such storage could continue for many more decades, given proper controls and supervision as well as repacking of some waste and periodic refurbishment of stores.

In recent years, there have been developments that have led some countries to consider whether the roles of storage might be expanded to provide longer-term care of long-lived solid radioactive waste and spent fuel. The consideration of such 'expanded roles' is linked to discussion of alternative strategies for the long-term management of long-lived solid radioactive waste and spent nuclear fuel, i.e. that final disposal is not necessarily the end-point or that it might only be implemented after an extended period of storage. In general, long-term storage involves packaging radioactive wastes and storing them in purpose-built facilities. Stores can be either above ground or below ground in the form of a single central facility or a range of local facilities. If above ground, they can be designed to withstand foreseeable attack. With periodic refurbishment, long-term interim stores might last for 100 years or more, depending on the design.

4.4.11 Waste transportation

Solid or solidified waste should be adequately packaged and contained for transport by road, rail, air or sea in accordance with the national legal

requirements. These national legal requirements should be based on the requirements established in Ref. [37] or in international agreements.

The on-site transport of radioactive waste may not need to meet all the requirements for off-site transport, because the shipment is at all times under the control of the operator, who is responsible for on-site operations. The operator should establish requirements and authorizations to ensure the safety of on-site transport. The facility operator should take into account in the site emergency procedures the possible exposure of a member of the public as a consequence of the on-site transport of waste, although such an exposure is unlikely.

4.4.12 Waste disposal

Disposal is emplacement of waste in an appropriate facility without the intention to retrieve, although some countries use the term *disposal* to include *discharges* of effluents to the environment [39].

Exempt waste is the waste that meets the criteria for clearance, exemption or exclusion from regulatory control for radiation protection purposes as described in Ref. [40]. Exempt waste is acceptable for disposal in landfill sites used for domestic and industrial waste.

Simple trenches have been used for many decades for the disposal of short-lived low and intermediate level waste. They are generally considered appropriate only for those wastes including disused sources that will decay sufficiently within an anticipated period of institutional control (generally between 100 and 300 years) to represent no risk to the public, as determined by safety assessments. The design and function of such repositories are described in Refs [41–44]. Large-scale (typically thousands of cubic metre capacity) near-surface engineered vault repositories have similar containment objectives and are used for similar types of RAW as simple trenches. Their engineering is intended to allow ease of waste emplacement and increased efficiency in the management and closure of the repository. The issue of post-institutional control intrusion can still be a dominant factor in waste acceptability [7, 11]. For the near-surface disposal option, a performance assessment is also required to determine either that the activity of the RAW can be contained until it has decayed or, if some migration is anticipated, that consequent doses are acceptable.

Near-surface shafts and/or boreholes can be considered as alternative or complementary to near-surface vaults. These disposal options have the advantages of being economical and also minimizing the probability of human intrusion. If necessary an engineered barrier system (EBS) can be added to the design and construction of these facilities to provide additional protection against radionuclide migration and human intrusion. More heavily engineered near-surface facilities have been designed with the

specific intention of reducing the likelihood of intrusion by emplacement of a massive concrete plug or cover over a large shallow shaft or borehole. For example, a reinforced concrete slab at least 1 m thick is considered to be a deterrent to inadvertent intrusion. These intrusion-resistant designs [45] will be helpful if institutional controls break down before the typically envisaged 300-year period. However, they do not offer a sufficient guarantee against intrusion to be considered for disposal of higher activity or longer lived waste.

Radioactive wastes that are not acceptable for disposal in near-surface disposal facilities, because they will not decay sufficiently within the period of institutional control, may be suitable for disposal at greater depth in disposal units characterized by one of several configurations [7]. At present, with the exception of deep tunnels and mines, it is uncommon to find construction work (e.g., deep foundation engineering) carried out at depths greater than about 30 m [44], so disposals at depths greater than this are only vulnerable to intrusion by deep drilling for water or mineral exploration – a much lower probability. As a result, the intrusion exposure risks posed by higher activity waste disposed of at intermediate depths are small. Shafts or boreholes to depths of several tens of metres or more are relatively simple to construct and can offer an attractive disposal option for small volumes of waste such as radioactive sources [44]. Evaluation of such options needs to consider the stability of the hydrogeological system over the time period of concern for containment, which may be several hundreds or thousands of years depending on the types of radioactive sources to be disposed of.

Very low permeability host rocks, with little or no advection of groundwater, can also provide adequate containment without the need for additional EBSs. Some clay and claystone formations at intermediate depths can provide such an environment, and evidence of lack of flow can be obtained from pore water environmental isotope analyses and evaluation of any fracturing that may be present in the rock. The isolation capability of this option depends on the ability to provide good shaft or borehole backfilling and sealing. The use of natural materials that reconstitute the original properties of the penetrated rock formations is recommended for all or some part of the sealing system. This may involve removal of some lining or casing to allow sealing against the host formations. If the disposal borehole/shaft is subject to significant water inflow or the geotechnical characteristics of the geological materials do not allow the excavation to be sufficiently stable, EBSs need to be emplaced to provide a level of containment commensurate with the hazardous life of the waste.

There are some disposal facilities for RAW in large rock cavities at depths of several tens of metres, generally in hard crystalline rocks such as granite (e.g., in Sweden and Finland). They are designed to contain short-lived low

and intermediate level waste. The containment provided by such repositories often comprises massive concrete vaults or silos, with additional EBSs such as clay backfills and buffers. This type of containment should be adequate for the disposal of many, if not all, types of RAW. For emplacement of high activity waste in a mined, intermediate depth repository, it is necessary to consider packaging and activity concentrations that suit the thermal characteristics of the host rock and EBSs of the repository. In addition, disused mines and/or caverns can be considered for intermediate depth disposal. Such facilities have not been widely used for the disposal of RAW. The objective of using deeper boreholes, at depths typical of geological repositories, would be to achieve greater isolation for limited volumes of RAW, including disused radioactive sources, in an environment that is characterized by lower flow, more stable chemistry and longer potential return paths to the biosphere, compared with the other options. In a very low permeability environment (e.g., some clay and claystone formations), there may be no effective water movement at depths of a few hundreds of metres. In such conditions, provided an adequate borehole seal can be constructed, containment of radionuclides is provided by the geological barrier and there is no requirement for supplementary EBSs beyond those needed to emplace the radioactive sources into the borehole and to maintain borehole stability during emplacement operations (casing and cementing). The option is particularly suited to the highest activity and long half-life radioactive sources, for which long containment periods are required (e.g., ~10–20 half-lives or more). For example, strong ^{226}Ra sources could require isolation for ~20,000–30,000 years. The depth and design of disposal also significantly reduces the likelihood of inadvertent intrusion, resulting in exposures to high concentrations of radionuclides before sources have decayed.

Mined repositories, comprising caverns or tunnels with varying types of EBSs, are being developed in many countries that have nuclear power industry wastes to manage [11, 46, 47]. They are designed to contain long-lived low and intermediate level waste, HLW and SFW. The containment provided by all such repositories is expected to be adequate for the disposal of all types of RAW provided that legal and regulatory requirements on repository inventory permit (some countries have strict constraints on the types of waste that can be placed in specific repositories which are purely legal and unconnected with safety and performance). In addition, disused deep mines and/or caverns could be considered for geological disposal [46, 47].

4.4.13 Disposal of NORM waste

NORM-containing waste is generally deposited in consolidated and overcovered piles or sludge beds, or purpose designed repositories with lined

cells and protective capping [28]. As it is not feasible to move such large amounts of material, the waste tends to be disposed of on the site of its generation. Capping and some engineered structures may be used to prevent erosion and to limit the leakage of radioactive gases. In some cases, the waste has been disposed of by using it to backfill disused underground mines. There is growing evidence to suggest that bulk wastes contained in properly engineered surface reservoirs have very low radiological impacts. However, their environmental, safety and financial liability implications can be seriously underestimated. This has been demonstrated in the case of phosphogypsum stacks, where recent developments have suggested that the stacking option is not optimal and that more attention should be given to beneficial uses of the material [28]. Landfill disposal has been demonstrated as being an appropriate option for dealing with many types of NORM residue for which the quantities and activity concentrations are moderate, including most types of furnace dust with enhanced concentrations of ^{210}Pb and ^{210}Po. Normal landfill facilities are generally suitable, but the presence of non-radiological contaminants such as heavy metals may require the use of landfill sites specially designated for hazardous waste. NORM residues from the chemical extraction of rare earths from monazite are produced in significant quantities and have characteristically high activity concentrations. It has been demonstrated that such wastes can be suitably disposed of either in earthen trenches or in engineered cells, depending on the activity concentration.

4.5 Methodologies for technology selection

The selection of pre-treatment, processing, storage and disposal technologies is necessarily bound up with the overall strategy for the management of the waste under consideration, and this is turn may be part of a larger scheme embracing many waste types. To achieve a satisfactory waste management strategy, waste management components must be complementary and compatible with each other [1]. Many aspects have to be addressed, the challenge being to achieve the optimal solution in a logical, structured and justified way. An existing IAEA publication has already reviewed most important factors affecting the selection and implementation of waste management technologies [48]. In any case, it is important to ensure that all three basic waste management routes (e.g., clearance, discharge or regulated disposal) are taken into account and evaluated in defining waste management strategies. It is important to consider strategies for all waste streams generated at facilities or sites rather than selecting options for individual waste streams. In addition, most national regulators now demand an impact assessment of proposed technologies and a justification for the selected technology.

The process of selecting a waste management technology typically starts by collecting and assessing available data, by considering all potentially influencing factors such as applicable regulations, waste properties, waste routes and associated good practice indicators. A set of possible technological options is then devised together with a preliminary waste management plan for implementing each option. These plans can be relatively brief at this stage but still sufficiently well defined, so that the associated major hazards and risks can be visualized.

The next step is to perform technology selection studies. During this process, formal decision-aiding techniques and 'workshop' discussion sessions can be employed. Selection of a preferred or optimized waste processing technology is best achieved through the evaluation of the general criteria and constraints in terms of their attributes for a specific waste stream or facility (Table 4.1).

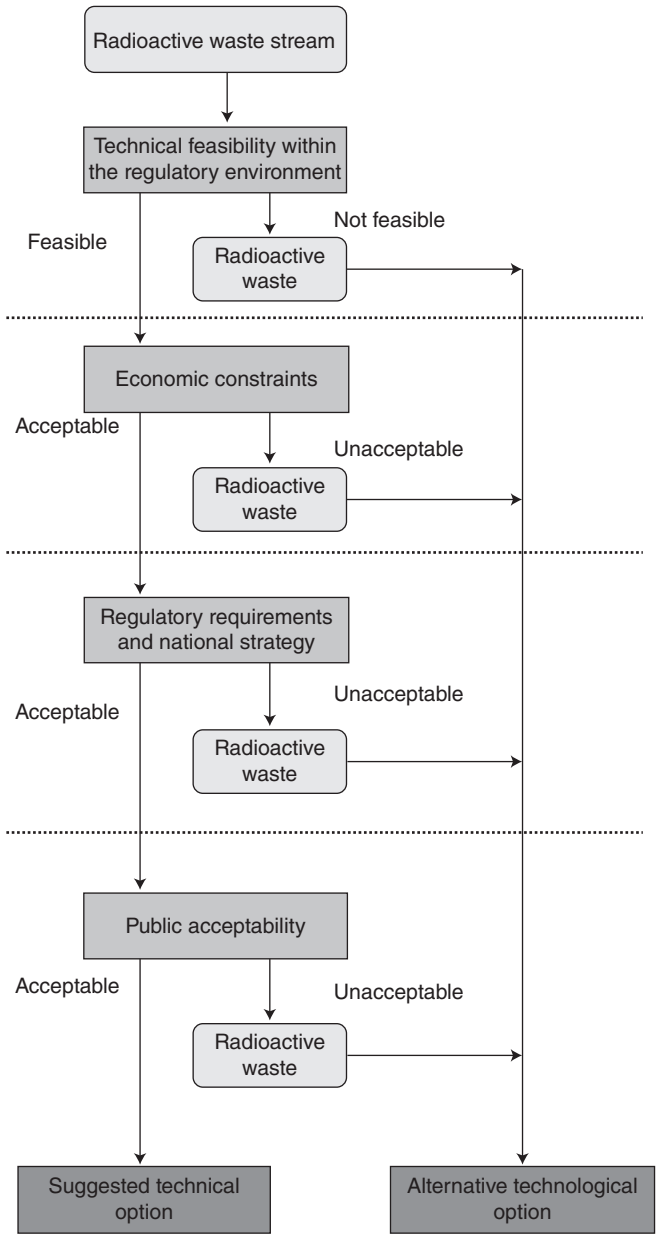
This evaluation can benefit from the use of formal decision-aiding techniques that address the influencing factors and associated good practice indicators. When evaluating the various influencing factors for a specific technology option, a simple 'decision-tree' approach could be adopted, in which the various factors are evaluated. The limitations of a linear approach are that influencing factors may only be considered one at a time, and in descending order of priority. Project selection decisions require multiple, generally non-linear, objectives to be simultaneously optimized. In addition, factors that are mutually influential cannot be considered in combination. An example of the simplified decision-tree approach for the selection of a suitable technology for a particular waste stream is given in Fig. 4.2, although not all criteria and constraints are accounted for.

4.5.1 Multi-attribute analysis

Multi-attribute utility (MAU) analysis [5] is a powerful tool accounting for many other criteria and constraints, besides costs, involved in the technology selection process. MAU analysis is an effective and efficient way of showing the impact of each technology option in terms of good practice attributes, and of reaching conclusions that address all of the influencing factors. Such analysis involves assigning numerical ratings and weightings to the factors considered, followed by comparison of the obtained total scores for the options. If necessary (i.e., when two options have very close scores), a sensitivity analysis can be performed to check whether or not the preferred option is the right choice. A simple scoring of the criteria for a given option allows any option to be discarded or considered for further evaluation. Regardless of the approach it is necessary to produce a justifiable and auditable solution for selected options.

Table 4.1 Technology-related criteria and attributes

Criteria		Good practice attributes
1	**National policy and strategies**	Compliance with the intent of national polices and strategies. In the case of insufficient national policies and strategies, compliance with international 'good practice'.
2	**Regulatory framework**	Compliance with the requirements of the regulatory framework. In the case of insufficient regulatory framework, compliance with international 'good practice'. Clearance levels are set up. Mechanism for authorized discharge is established.
3	**Funding and cost**	Both direct and indirect costs (e.g., stakeholder involvement and public acceptance) addressed. Total cost of the viable technology evaluated or compared and technology selected/eliminated in terms of main cost factors. Adequate financial resources or financial security and funding mechanisms available for the funding of viable technology.
4	**Health, safety and environmental (HSE) impact**	HSE impacts of viable technologies known and considered in the selection of technologies; HSE impact optimized by reducing exposure of the workforce and members of the public. The need for transportation of radioactive material is minimized.
5	**Waste characterization**	Identification of all sources of waste generation. Waste characterization developed and can be implemented at all stages of the waste management process.
6	**Waste management system**	Waste management system exists and can support the newly introduced technology. Storage/disposal facilities available. Operational waste generation control programme in place.
7	**Human resources**	Availability of suitably qualified and experienced personnel. Consideration of lessons learned from implementation of other technologies.
8	**Social impacts and stakeholder involvement**	Technologies discussed with stakeholders and considered in a transparent way. All stakeholders involved in the selection of a technology and reasonable consensus reached.
9	**Technical factors**	All technical factors affecting the selection of a technology (e.g., maturity, robustness, complexity and maintainability, etc.) are taken into account.
10	**Physical infrastructure**	Physical structure is available and can support the newly introduced technology.



4.2 Linear decision-tree approach for the technology selection of a waste stream.

The MAU method makes clear to all involved the basis on which the alternatives are being evaluated. It offers quantifiable principles for choosing options. This is particularly important in group decision-making situations, in which many different points of view and alternative decisions have to be reviewed and taken into account. The attributes needed for evaluation of options must be identified. They are assigned a weight that reflects their importance to the decision. A value of 3, 2 or 1 might be assigned to each attribute, depending on its importance. Alternatively 100 points can be assigned and distributed over the attributes according to their importance. A score can be given to each of the alternatives for each attribute. A scale of 1–10 may be used. The score of each alternative for each attribute is then multiplied by the weight of that attribute, and the total is calculated. That total represents the value of that option, which can then be compared to the same calculation for the other options. If it is a group process, each member of the group scores the attributes for each option and the group's ratings can be totalled or averaged. The final result of this example analysis would be a relative, numerical ranking of the options (the score for each option).

Furthermore, various criteria such as non-safety-related matters could also be considered in the process of selecting an option. Where relevant, safeguard-related issues should also be considered in optimizing both safety and resources in the decision-making process. The costs of maintenance, surveillance and physical protection for the waste management facility should also be taken into account. It should be ensured that the selected option meets all the applicable safety requirements. A MAU model can be used to further explore the consequences of changing the attributes, their weights, or the scores they received. Since the criteria are transparent, it is possible to make several changes and review the results. For example, if it appears that some attributes are too important in determining the results, the weights could be adjusted to produce more realistic results.

Workshop sessions (sometimes called brainstorming sessions or decision conferences) can provide a practical and motivating way forward. In such sessions a panel of relevant experts (including experienced operators) agree on the list of influencing factors and then assess the impact of these factors on each of the technological options, assisted by the use of decision-aiding techniques. It is important to produce a report of the workshop sessions, describing the technique adopted, the considerations addressed and the results obtained. This report can be a valuable aid in support of the waste management plan and the associated safety justification.

The processes of selecting a preferred technology and the subsequent detailed strategy are best approached by ensuring that the team clearly understands the underlying safety logic. This logic must be applied to each of the candidate options (at an appropriate level of detail), as part of the

process of selecting a preferred option. The key point is to ensure that there is a demonstrable connection between the characteristics and amounts of radioactive waste at generation, the proposed technologies, the associated risks in implementing these technologies, the safety management arrangements, and costs. For example, analysis of the risks involved logically determines the requirements for key aspects such as additional or modified equipment, staff training, procedures, work instructions, maintenance and security arrangements.

4.6 Conclusion

Before embarking on the selection of a particular technology or selection of options to address complex waste management needs, it is essential to analyse the waste generation thoroughly and understand the properties, types and volumes of waste. It is necessary to fully comply with the regulatory regime, and to ensure that disposal options are available. It is assumed that legal and regulatory infrastructure related to waste management exists or is going to be established as soon as practicable. The selection of a technology/technical option needs then to be based on the evaluation of all relevant criteria and constraints.

More details on this topic will be provided in a forthcoming IAEA publication that is tentatively titled 'Selection of Technical Solutions for the Management of Radioactive Waste'.

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5

Irradiated nuclear fuel management: resource versus waste

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Abstract: Management of irradiated fuel is an important component of commercial nuclear power production. Although it is broadly agreed that the disposition of some fraction of the fuel in geological repositories will be necessary, each of the fuel cycle options (once-through, limited recycle, advanced partitioning and transmutation, fuel breeders) introduces distinct waste management challenges. The choice of options significantly affects what fraction of material will be disposed in what manner. To further complicate this question, until geological repositories are available to accept commercial irradiated fuel, these materials must be safely stored. This chapter discusses some of these options.

Key words: nuclear fuel cycle, Yucca Mountain, waste forms, separations, partitioning.

5.1 Introduction

Nuclear power accounts for 22% of global electricity generating capacity (NEA, 2010). It is anticipated that nuclear power will continue to supply approximately 20% of the world's electricity through at least 2035. According to the World Nuclear Association, 'A typical 1000 MWe light water reactor will generate (directly and indirectly) 200–350 m^3 low- and intermediate-level waste per year. It will also discharge about 20 m^3 (27 tonnes) of used fuel per year, which corresponds to a 75 m^3 disposal volume following encapsulation if it is treated as waste. Where that used fuel is reprocessed, only 3 m^3 of vitrified waste (glass) is produced, which is equivalent to a 28 m^3 disposal volume following placement in a disposal canister' (World Nuclear Association, 2011). These estimates will vary depending on the degree of separation applied in the reprocessing and other factors. The worldwide inventory of existing (as of 2009) irradiated fuel resulting from nuclear power production is 165,390 metric tons, and the annual production rate in 2009 was approximately 7,300 metric tons (NEA, 2010). This rate of irradiated fuel production is expected to continue (and in all likelihood will

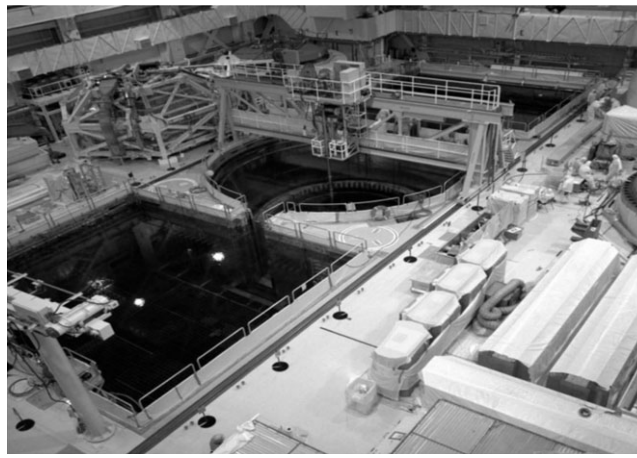
increase) through 2035. Managing this highly radioactive material poses significant challenges to the nuclear power industry and to human society.

Emplacement in deep geological repositories is commonly accepted as the best option for disposing of the waste components of irradiated fuel. However, no repositories suitable for accepting irradiated fuel from power reactors exist. Furthermore, establishing the location of geological repositories is difficult both technically and politically, as illustrated by the US experience with the planned repository at Yucca Mountain in Nevada. Thus for the foreseeable future, utilities will continue to store the irradiated fuel at the reactor sites. There is also some debate as to whether irradiated power reactor fuel should be viewed as simply waste (as in the once-through fuel cycle), or as a resource (closed or partially-closed fuel cycle). This chapter will address these issues as they relate to the management of commercial irradiated (also termed spent or used) fuel.

5.2 Temporary storage

At all nuclear power reactors, irradiated fuel is first transferred from the reactor into water-filled storage pools. The fuel is stored in geometric arrangements that eliminate the possibility of an inadvertent criticality. Over the years, as irradiated fuel has continued to accumulate, utility operators have implemented storage technologies that have increased the capacity of the storage pools, without increasing the size of the pools themselves. This has been achieved by re-racking the fuel bundles into storage arrays that incorporate neutron absorbers. Criticality control through adding soluble boron to the pool and through placement of neutron-absorbing storage rack inserts have further increased pool storage capacity. These technologies have made it possible to space the fuel in the storage pool at nearly the same packing density that is achieved in the reactors themselves (Kessler, 2010). Figure 5.1 shows a typical pool for storage of irradiated fuel.

Despite increases in pool storage capacity, utilities are continuing to move towards adding dry storage capacity. This need is driven principally by continuing delays in the establishment of geological repositories, but the experience with pool-stored fuel at the Fukushima Daiichi nuclear complex following the earthquake/tsunami event might also provide impetus to more quickly move irradiated fuel to dry storage. Although early development of dry storage technologies was directed simply at fuel storage, the approach has evolved into developing containers that serve both as a storage container and as a shipping cask, so-called dual- or multi-purpose dry storage systems. The advantage of this approach is that the fuel, once removed from the storage pool, has to be handled only once; since the storage container and shipping cask are one and the same, there is no need to move the fuel from the storage container to a suitable shipping cask. A



5.1 Example of a fuel storage pool. This photo was taken during transfer of fuel from the reactor core (center) to the fuel storage pool (left). Courtesy of Energy Northwest, Richland, WA.

5.2 Dry fuel storage facility with reactor building in the background. Courtesy of Energy Northwest, Richland, WA.

number of dual-purpose dry storage containers have been developed and these have been well described elsewhere (Kessler, 2010). Figure 5.2 illustrates a typical dry fuel storage facility.

5.3 Fuel cycle options

The byproducts of the operation of fission reactors can be managed in two ways: without treatment that supports recycle or with the recycle option

enabled. In the former approach (once-through or open cycle), the fissile resource is the ^{235}U taken from the ground (enriched to 3–5% to allow light water moderation of neutrons) and the several percent of ^{238}U that is transmuted to ^{239}Pu and fissioned during normal reactor operations. The used fuel once removed from the reactor is considered as waste and is managed accordingly. The latter option (closed loop) can be pursued with varying degrees of processing, though at present only plutonium isotopes are recycled in mixed oxide (MOX) fuel (while the residual $^{235,236,238}U$ is stored for future use).

Though plutonium production reactors and reprocessing facilities operated within the nuclear weapons complex in the US between 1944 and the early 1990s, the reprocessing of spent fuel from commercial reactors was practiced for only a very brief period during the 1970s. A *de facto* moratorium was placed on reprocessing of commercial spent nuclear fuel in the US in 1977; this ban was lifted in 1981, but no attempts were made to revisit this option until recently. A 2005 energy bill has again allowed consideration of more complete used fuel management and has spurred a modest revival of research into reprocessing (and transmutation) options. However, current US nuclear fuel management policy remains the once-through option with direct disposal in a deep geological repository.

Most nuclear power producing nations practice the once-through option; France, Japan, the UK and Russia operate at least partially closed fuel cycles in which fuel grade Pu is recovered for recycle. At present, no country operates a more complete recycling program, though research exploring options is in its third decade. It can easily be understood that the waste management issues associated with these options are markedly different (although ultimately it is generally accepted that *every* option will require a geological repository for the residual radioactive materials).

5.3.1 Once-through

In the 'once-through' fuel cycle concept, the 3–5% ^{235}U enriched fuel is burned once in the power reactor. It is then removed from the reactor, stored temporarily, and ultimately packaged for disposal in a deep geological repository. Because the once-through fuel cycle has been the official policy for managing irradiated commercial nuclear fuel in the US, the geological repository program in the US was highly developed. It had advanced to the point that the US Department of Energy (DOE) submitted the license application for the Yucca Mountain repository to the US Nuclear Regulatory Commission in 2008 (OCRWM, 2008). Although the DOE recently withdrew the license application for the Yucca Mountain repository, the development of this repository represents a good example

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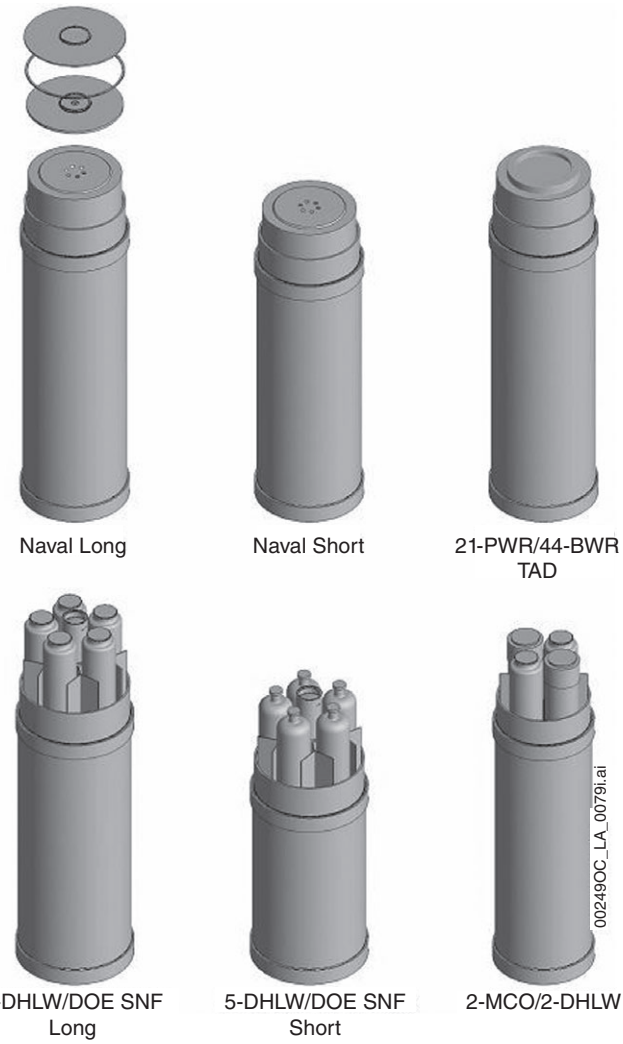
appropriate for the once-through fuel cycle. For this reason, the discussion in this section will focus on the Yucca Mountain repository concept.

The site of the proposed Yucca Mountain repository is located in an arid region, about 145 km northwest of Las Vegas, Nevada. The geology and hydrology of this site have been extensively studied, as have the volcanic and seismic characteristics of the area (OCRWM, 2002). The Yucca Mountain repository is designed to accommodate three primary waste types: (1) commercial irradiated nuclear fuel from power generating plants, (2) DOE-owned irradiated fuel, including naval reactor fuel, and (3) vitrified high level waste (HLW). The latter category consists mostly of HLW borosilicate glass canisters generated from vitrifying radioactive tank wastes at the Hanford and Savannah River sites in the US (see Chapter 18). Under current US law, a maximum of 70,000 metric tonnes heavy metal (MTHM) can be disposed of at the Yucca Mountain repository. Of this, 63,000 MTHM is allocated to the disposition of commercial irradiated fuel, representing approximately 221,000 fuel assemblies (OCRWM, 2008). The DOE-owned irradiated fuel would constitute 2,333 MTHM, and the remaining 4,667 MTHM would be allocated for disposal of vitrified HLW. It is interesting to note that under the current statute, the Yucca Mountain repository (if built) would already be oversubscribed because the 4,667 MTHM allocated to vitrified HLW represents only approximately 9,334 of the 22,000 canisters expected to be produced and also 292,000 commercial irradiated fuel assemblies are expected to be produced by 2040, far in excess of the space allotted for 221,000 assemblies (OCRWM, 2008).

There is a single design for the Yucca Mountain waste package, but it has six configurations providing flexibility to accommodate the different types of waste to be received. Figure 5.3 illustrates the six waste package configurations (OCRWM, 2008). All six configurations consist of two concentric cylindrical containers. The primary (inner) waste container is made of 316 stainless steel and has walls 50.8 mm thick (Skinner *et al.*, 2005). The outer secondary containment has 20.3 mm thick walls and is made of alloy C-22. The secondary container is designed to provide corrosion resistance, so it is referred to as the outer corrosion barrier. Each waste package has three welded lids, one on the primary container and two on the corrosion barrier. After welding of the inner lid, the primary waste vessel is evacuated and back-filled with helium. The helium serves three primary purposes:

1. It inhibits internal corrosion.
2. It improves heat transfer between the waste and the waste package.
3. It provides a tag for leak testing of the inner vessel closure welds.

Because of the intense radiation involved, all the waste package closure operations must be conducted robotically (Skinner *et al.*, 2005).

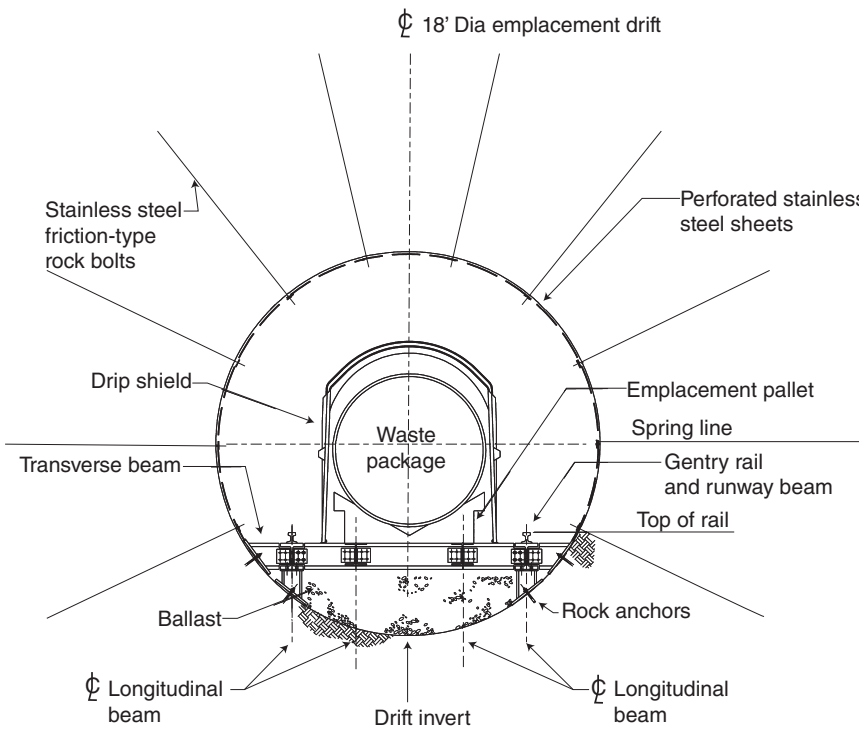


5.3 Approved waste package configurations for the Yucca Mountain repository.

In the design concept for Yucca Mountain, the loaded waste packages were to be transported to emplacement drifts carved within the repository. The emplacement drifts are nominally 5.5 m in excavated diameter with an average length of approximately 600 m. The length of the emplacement drifts is constrained to no more than 800 m to allow for efficient ventilation. The drifts have been excavated in parallel, spaced 81 m apart. This spacing is designed to prevent thermal interaction between adjacent drifts and allows infiltrating water from the surface to percolate past the drifts.

Encroachment of water onto the waste packages was to be further mitigated by installation of a titanium drip shield. Water percolating into the emplacement drift from above the waste package is directed to a point below the waste package by the drip shield. Figure 5.4 provides a cross-sectional illustration of an emplacement drift, as designed for the Yucca Mountain Repository (OCRWM, 2008).

Sweden also practices a once-through nuclear fuel cycle and substantial progress has been made in that country on the establishment of a geological repository (see Chapter 13). These efforts are led by the Swedish Nuclear Fuel and Waste Management Company (SKB). Three decades of research and development and a 20-year site development process has resulted in the selection of Forsmark in the municipality of Östhammar as the site for the Swedish geological nuclear repository. In contrast to the arid environment of Yucca Mountain, the Forsmark site is located along a coastal area. However, there are relatively few water-conducting fractures in the bedrock at the depth of the fuel emplacement (500 m) at the Forsmark site (SKB, 2009). For emplacement into the Swedish repository, it is planned that the irradiated assemblies will be packed into cast iron baskets, which



5.4 Cross section of the Yucca Mountain emplacement drift.

in turn will be placed within thick copper canisters. Once placed within the repository, the loaded copper canisters will be packed in bentonite clay.

Finland is taking a similar approach to Sweden, having chosen the Olkiluoto site for the Finnish HLW repository (Okko and Rautjärvi, 2004). In this case, the waste packages will be placed in excavated tunnels hundreds of meters below the surface. The tunnels will be separated by a distance of 25 m. As for the Swedish repository, the irradiated fuel will be packaged into nodular cast iron containers, which will then be enclosed in a 5 cm thick copper shell. The waste packages will be surrounded with bentonite clay to absorb water and to protect the waste from minor movements in the surrounding bedrock.

5.3.2 Plutonium uranium recovery by extraction (PUREX) and mixed oxide (MOX)

The history of recycling of used nuclear fuels traces its lineage back to the 1940s and the Manhattan Project (Rhodes, 1986). Having begun with co-precipitation technologies, the multiple advantages of solvent extraction-based processing significantly improved the efficiency and throughput of recycling. Introduction of solvent extraction methods also dramatically reduced the amount of HLW produced during recycling operations. During the first several decades of this enterprise, the principal driver for reprocessing was plutonium production to support the weapons programs of the nuclear nations. Dating from the mid-1950s, the system of choice for plutonium isolation and recovery from dissolved used fuel has been the PUREX (plutonium uranium recovery by extraction) process. When introduced, the PUREX process represented a significant leap forward in efficiency over the solvent extraction processes that it replaced (REDOX and BUTEX; Nash *et al*., 2006).

In PUREX, contact of a 3–6 M HNO_3 solution containing all but the most volatile products of fission with an immiscible kerosene solution of tributyl phosphate (TBP) allowed selective extraction of $UO_2(NO_3)_2$ and $Pu(NO_3)_4$ from a mixture of fission products and mixed actinides. With careful control of the conditions of extraction, all fission products, the transplutonium actinides and neptunium remain in the aqueous raffinate for ultimate disposal as high level wastes. Operation of the solvent extraction system with multiple stages and a counter-current flow of aqueous and organic phases results in a high purity Pu/U nitrate organic solution almost completely devoid of fission products or minor actinides (Np, Am, Cm).

From this solution the plutonium is readily (and selectively) removed with the application of a suitable reducing agent (early applications employed Fe(II) sulfamate, more recently U(IV) or HN_2OH/N_2H_4 have been employed as reducing agents). The resulting pure Pu product is well

suited to further purification to the metallic state or the creation of MOX ceramic fuel in combination with fresh UO_2. Most of the uranium is removed from the TBP phase by reducing the acidity; the remainder is scrubbed with complexants to allow the extractant to be recycled back to the head end of the process. Because the isotopic distribution of the uranium is altered during its time in the reactor, this material is not at present recycled back to the MOX fuel, but rather stored for future use. The PUREX process has seen more than 50 years of process improvements and remains today the standard for Pu/U recycle. Various adaptations of this extraction system include options for extraction of Np(VI or IV) and the preparation of mixed U/Np/Pu mixtures to increase the proliferation resistance of the fissile material.

5.3.3 Advanced partitioning and transmutation

The long-lived radioactive components of irradiated nuclear fuel serve to create a monumental challenge for managing the wastes from operation of nuclear power reactors. In the open fuel cycle option, the presence of alpha-emitting ^{239}Pu (0.5%, $t_{1/2}$ = 24,100 yr), ^{237}Np (0.05%, $t_{1/2}$ = 2,100,000 yr), and ^{241}Am ($t_{1/2}$ = 432 yr, low primary yield, but of increasing importance with time in storage as the daughter of the β^- decay of ^{241}Pu – 0.1%) demands that a geological disposal system must ensure retention of these isotopes within the repository environment for 240,000 years or more. Even with partial recycle (recovery of Pu isotopes to MOX fuels), the residues of Am and Np isotopes leave a considerable challenge for repository development. The need to qualify a repository for a span of time longer than that of all human civilization has been used to support arguments for further treatment (beyond PUREX processing) of dissolved nuclear fuels to enable transmutation of these elements in advanced reactors or by accelerator transmutation.

With the PUREX process, or an equivalent U/Pu management scheme that addresses concerns about the proliferation of nuclear weapons, the challenge of advanced nuclear fuel cycles to support transmutation of minor actinides ultimately focuses on the isolation of Np, Am and perhaps Cm from fission products. Of these, the easiest task is control of the partitioning of Np. In PUREX processing, Np can be made to partition to either the aqueous or organic phase based on its oxidation state. In HNO_3 media less concentrated than about 4 M, NpO_2^+ is the dominant species, which is poorly extracted by TBP, hence Np remains with the fission products. At higher concentrations of HNO_3, NpO_2^+ is susceptible to oxidation to the hexavalent state (NpO_2^{2+}), which readily co-extracts with UO_2^{2+}. Other options feature the introduction of reducing agents that convert NpO_2^+ to extractable Np^{4+}. In modern adaptations of PUREX (e.g., UREX, COEX),

mixtures of U/Np/Pu are created to increase proliferation resistance of the products.

Once choices are made regarding the partitioning of Np and Pu in PUREX/UREX/COEX processing schemes, the remaining challenge is recovery (for transmutation) of Am, with or without Cm. Following PUREX/UREX, Am and Cm remain with the fission products in the raffinate. Am and Cm are most stable in acidic solutions as the trivalent cations, which is also the common oxidation state of the fission product lanthanides. This coincidence of stable oxidation states and limited reduction/oxidation (redox) adjustment options is unfortunate, as the trivalent actinides and lanthanides are also of similar size, and thus quite difficult to separate. Their mutual separation is an important issue, as some members of the lanthanide series have high neutron capture cross sections, thus they compete for the neutrons that might otherwise transmute actinides.

However, the tendency of cation radii to decrease across both series, combined with the slightly softer acidic nature of the actinide ions compared to the lanthanides in terms of the hard-soft acid-base theory (Pearson, 1963), offers an avenue to an aqueous processing pathway to support this separation and transmutation goal. The simplest solution to the separation of trivalent actinides from fission products is to develop soft donor extractant molecules that selectively remove trivalent actinides from the fission product mixture (i.e., rejecting the lanthanides). Unfortunately, soft donor extractants based on N or S donor functional groups have much higher affinity for selected transition metal fission products (noble metals) and corrosion products (Fe, Ni, Co, Zn); as a result a single-step isolation of the trivalent actinides from this mixture has thus far proven elusive. Advanced fuel cycle research efforts have as a result evolved toward a two-step process of extracting trivalent lanthanides and actinides away from the remainder of the fission residues followed by the mutual separation of the f-elements. The current state of this art has been described in detail previously (Nash *et al*., 2006), hence will be summarized only briefly here.

Motivated by the need to clean up residual wastes within the US nuclear weapons complex, research into the development of new reagents for total actinide removal from dissolved acidic waste solutions (similar to PUREX raffinates) arguably began with studies of extractants combining phosphine oxide and amide functional groups in the same extractant molecule. Based on the work of Siddall (1958, 1963, 1964), Horwitz and co-workers developed a family of carbamoyl(methyl)phosphine oxide extractants designed to follow PUREX with selective partitioning of trivalent f-elements (along with any residues of tetra- and hexavalent actinides); the TRUEX process was developed based on these reagents (Schulz and Horwitz, 1988).

With the aim of eliminating the use of organophosphorus extractants from advanced reprocessing schemes, French researchers followed with the

demonstration of similar separation systems based on tetra-alkyldiamides of malonic acid (malonamides; Madic *et al*.,1994), which performed similarly from more acidic media and with fewer complications arising from extractant degradation products. Sasaki and co-workers (Morita *et al*., 2002; Sasaki *et al*., 2001) subsequently produced tetra-alkyl diamides of diglycolic acid (e.g., tetraoctyldiglycolamide, TODGA) that introduced the interesting feature of maximum extraction of trivalent actinide nitrates under some conditions. In China, Zhu and co-workers developed a similar system based on commercially available trialkylphosphine oxide extractants (Zhu *et al*., 1983; Zhu and Song, 1992). This process required operation at reduced concentrations of HNO_3, but similarly resulted in selective partitioning of trivalent actinides and lanthanides away from PUREX raffinates. Fuel cycle research continues to examine the potential for application of each of these classes of reagents.

With the f-elements thus separated from the problematic (from a separations perspective) transition elements, attention turns toward the mutual separation of trivalent lanthanides and actinides. Several different approaches to pyrometallurgical separations offer electrochemical pathways to a partial separation of the groups based on electrodeposition (Nash *et al*., 2006). Though research continues on both aqueous and pyrometallurgical methods, it is clear at present that more efficient separations can be achieved using wet methods (at the cost of increased secondary waste volumes that are inherent to aqueous techniques). Both soft donor extractants and water soluble holdback reagents to selectively prevent actinide extraction have been studied for the purpose of separating these groups, each with some degree of success, but also with limitations. Though some have been characterized through the pilot stage using actual dissolved used fuel, to date none have been implemented on an industrial scale. The most mature options are described in brief in Table 5.1.

In the partial recycle options, U, Np, Pu mixtures are the products that must be managed for their possible recycle in (first) light water and (subsequent to the first cycle) advanced reactors. The wastes from this option that require disposal in a geological repository include most fission products (noble gases and potentially other volatile fission products like iodine would be managed separately) and the transplutonium actinides (Np, Am, and Cm). The likely form of this waste would be some formulation of borosilicate glass. Repository retention times would be determined principally by the decay/release profiles of ^{241,243}Am, ^{129}I, and ^{99}Tc. In the more advanced fuel cycle options involving actinide transmutation, long-term radiotoxicity is reduced through the transmutation of Am, but ^{129}I and ^{99}Tc remain a long-term concern. The radiotoxicity of residual ^{129}I and ^{99}Tc is considered to be less than that of the original uranium ore; however, the environmental mobility of these isotopes remains a concern. The radiotoxicity is reduced

Table 5.1 Summary of aqueous processing options for separation of minor actinides from fission product lanthanides

Process	Comments	Status
TALSPEAK	DTPA complexes An(III) and HDEHP extracts Ln(III) from acidic streams.	Hot cell demonstration
'Reverse' TALSPEAK	An(III) and Ln(III) are both extracted. DTPA (and a carboxylic acid buffer) is then used to strip An from Ln	Complete partitioning system demonstrated with dissolved spent fuel in the CTH process
DIDPA	Modified reverse TALSPEAK. DIDPA used as the extractant, DTPA to strip Am, Cm.	Hot cell demonstration with actual waste
SETFICS	Modified TRUEX process. Uses DTPA complexant and modified CMPO extractant.	Not tested with actual waste
PALADIN	Malonamide co-extracts An(III) and Ln(III) from acid stream. HDEHP extractant and DTPA complexant selectively strips actinides.	Successfully tested
SANEX processes	Selective extraction of An(III) from Ln(III): Cyanex 301 uses R_2PSSH as extractant. ALINA uses two extractants: a dithiophosphinic acid and trioctylphosphine oxide. BTP/BTBPs and TMAHDPTZ w/ octanoic acid have been used as neutral extractants.	Demonstrated with Am, Ln mixtures and genuine wastes
DIAMEX-SANEX, GANEX,	Combined diamide/cation exchanging (HDEHP) extractant with selective stripping of trivalent actinides using buffered solutions of aminopolycarboxylic acid complexants.	Demonstrated with Am, Ln mixtures and genuine wastes

DTPA – Diethylenetriaminepentaacetic acid, DIDPA – Diisodecylphosphoric acid, CMPO – Octyl. Phenyl-N,N-diisobutyl carbamoylmethyl phosphine oxide, HDEHP – Di-2-ethyl(hexyl)phosphoric acid, BTP/BTBP – bis-1,2,4-triazinyl pyridine/bipyridine, TMAHDPTZ – 4,6-di-(pyridin-2-yl)-2-(3,5,5-trimethylhexanoylamino)-1,3,5-triazine.

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to less than the uranium ore after the passage of ten half-lives of ^{137}Cs and ^{90}Sr.

5.4 Managing wastes from fuel recycling

Implementation of any industrial operation to recycle one or more components from irradiated fuel will result in the generation of a number of waste streams that must be properly managed to mitigate environmental consequences. The primary wastes from aqueous reprocessing of spent fuel include cladding hulls and hardware, undissolved solids, gaseous fission products, and HLW raffinate. A host of job control, maintenance, and operational secondary wastes (typically low or intermediate level) are also generated. In this section, the nature of these waste streams and the methods in which they can be immobilized for disposal are summarized.

5.4.1 Hulls and hardware

Stainless steel hardware (including spacers, clips, springs, end plates, etc.) is removed in the first stage of reprocessing. The primary radionuclides in the hardware come from activation of stainless steel components (e.g., ^{54}Mn, ^{55}Fe, ^{60}Co, and ^{63}Ni). This stream makes up roughly 5 wt% of the fuel assembly. Cladding (sometimes referred to as hulls) refers to the metal tubes used to hold the fuel. At present, cladding compositions are typically >95% Zr with Sn and/or Nb alloying agents although stainless steels are sometimes used (e.g., in UK advanced gas-cooled reactor fuel). The primary radionuclides in and on the cladding are activation products (e.g., ^{95}Zr, ^{95}Nb, ^{54}Mn, ^{55}Fe, ^{60}Co, and ^{125}Sb), tritium, and transuranics/fission products from alpha recoil at the fuel–cladding interface. Tests have shown that the transuranic contamination resides in the inner 7 μm of the cladding. As cladding makes up roughly 25% of the used nuclear fuel mass, it is the largest single fuel component after UO_2.

Traditionally, hulls and hardware have been managed together. The most common approaches to managing these wastes are to wash and then (1) embed them in cement for disposal, (2) dispose directly, and (3) compact and dispose (IAEA, 1985). Compared to direct disposal, the compaction reduces disposal package volume by roughly a factor of four, while encapsulation in cement increases the volume by roughly 100% (double the volume).

A number of alternative approaches have been studied for these wastes including: rolling compaction and cementation, embedding in graphite, compaction with malleable metals (e.g., Pb), compaction and encapsulation in low temperature metals, powder metallurgical encapsulation, glass encapsulation, cryogenic crushing and encapsulation, oxidation and conversion

to ceramic waste forms (e.g., zircon), oxidation and cementation, hot pressing, melting to a Zr-Fe alloy zirconium separation using reactive gasses (Collins *et al.*, 2011). Although these methods are not being currently implemented, many show promise for improved waste management compared to the reference technologies.

5.4.2 Fission gasses

Almost all of the radioactive gasses (H, C, Kr, Xe, Cl, and I) are released during voloxidation[1] (if performed) and dissolution. They are carried in a relatively dry stream from the voloxidizer if present or in a wet stream containing significant nitrogen oxide species from the dissolver. These gaseous components can be selectively captured for immobilization. In the US, Cl and I must be captured for virtually all fuels and Kr must be captured for fuels cooled less than 30 years and ^{3}H from fuels cooled less than ~50 years. Figure 5.5 shows an example of the gaseous fission products evolved from various fuel recycling steps. Of primary interest are ^{3}H, ^{129}I, ^{14}C, and ^{85}Kr.

Tritium removal from voloxidizer off-gas is performed using a desiccant (such as $CaSO_4$) or a molecular sieve (such as Linde type 3A). The water content of air fed to the voloxidizer is controlled to obtain the desired decontamination factor in the tritiated water removal bed without significant increases in the tritium waste stream volume. Capture is performed near room temperature followed by release of the tritiated water at higher temperature. The captured $(H,D,T)_2O$ is then immobilized for decay storage and disposal. With a 12.3 year half-life, tritium immobilization does not require a robust waste form. Current process development activities assume that the tritium waste form is sufficiently low in long-lived radionuclides to qualify for near-surface disposal and the target waste form is generally considered to be a low-water cement. The other leading candidate is cementation of the loaded sorbent.

Iodine-129 and ^{36}Cl are significant dose contributors for nearly all repository environments because they are highly mobile, have long half-lives (15.7×10^6 years and 0.3×10^6 years, respectively), and are efficiently concentrated in the human body. Therefore, every reprocessing nation has strict tolerances on the capture of ^{129}I at a minimum. Various past studies have shown

[1]Voloxidation is a potential process step employed primarily to remove ^{3}H from the fuel meat prior to dissolution so that waste streams from all downstream solvent extraction processes are not ^{3}H contaminated. Tritium capture may not be necessary in all countries to the same level as required in the US (40CFR61 and 10CFR20), so processes with and without voloxidation will be considered.

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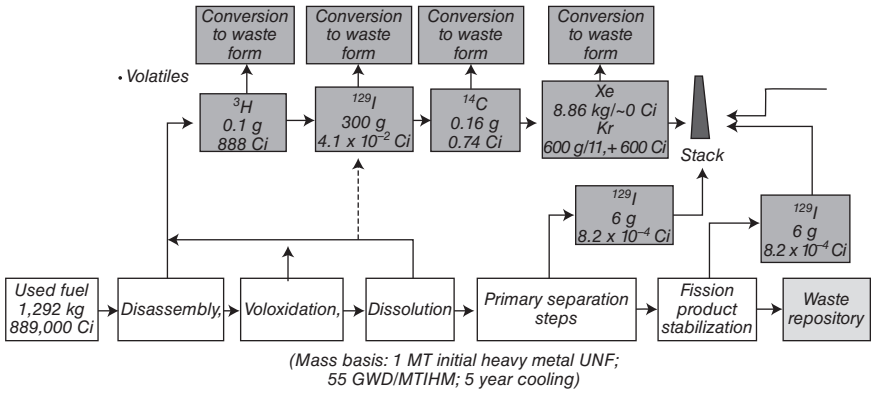


5.5 Schematic of off-gas treatment components from a typical UNF (per kg initial U after 55 GWd/MTHM and 5 years of cooling) (Jubin *et al.,* 2009).

that 94–99% of iodine reports to the dissolver off-gas. A large fraction of the iodine in the off-gas was found to be associated with organic compounds (e.g., methyl iodide). A range of technologies have been employed to capture iodine from the plant off-gas streams including (IAEA, 1980):

- silver saddles ($AgNO_3$ on ceramic substrate) → Hanford and Savannah River
- silver faujasite (AgX) → Sellafield
- silver mordenite (AgZ) → Hanford
- $AgNO_3$ on silica (e.g., AC-6120) → WAK and Mayak
- silver on alumina → LaHague, Rokkasho
- carbon → Hanford
- wet caustic scrub (2 M NaOH) → La Hague, Tokai, Krasnoyarsk, Mol, and Sellafield
- IODOX (20+ M HNO_3)
- mercurex (mercuric and nitric acids) → Dounreay and West Valley
- cadmium faujasite (CdX)

Advancements in materials science have allowed for the development of improved solid getter materials for iodine. Chief among them are silver-loaded aerogels (Strachan *et al*., 2010a; Matyas 2012); metal organic frameworks (Nenoff *et al*., 2011; Sava *et al*., 2011) and chalcogenide-based glass aerogels (chalcogels) (Strachan *et al*., 2010a). However, these materials are currently in the development phase and are not ready for full implementation.

Iodine waste form development and waste management are closely coupled to the separations technique employed. For example, at La Hague

in France and Sellafield in the UK, iodine is managed by ocean disposal (isotope dilution) which leads to the most appropriate capture method of caustic scrubbing. Other than ocean disposal, the immobilization/management of iodine is still a significant technical challenge faced by the industry in general. Several waste forms have been proposed and are being developed for the disposal of radioiodine.

Silver-loaded adsorbers (AgZ, AgX, AC-6120, alumina, etc.), for example, can be encapsulated in cements (Toyohara *et al*., 2002; Scheele *et al*., 2002) or low melting metals (Vance *et al*., 2005) or glasses (Garino *et al*., 2011; Perera *et al*., 2004), or hot pressed into a durable waste form (JAEA, 2007). Scheele *et al*. found that adding CaI_2 to the grout would significantly reduce the leaching rate of ^{129}I by isotopic dilution in the pour water solution (Scheele *et al*., 2002). However, for some repository design concepts, the presence of cement is a disadvantage because of the impact of alkaline cement leach solution on the corrosion of HLW glass and SNF. For example, the Yucca Mountain repository design specifically excluded cement wherever possible. The loaded AgI containing ceramics or glass can be hot-pressed into a final waste form (Sheppard *et al*., 2006).

Alternatively, the iodine can be eluted from the capture media and immobilized. Pure halide waste can be immobilized in:

- bismuth oxide-based ceramics (Krumhansl and Nenoff, 2011)
- sodalite-like minerals (Strachan and Babad, 1979; Winters, 1980; Nakazawa *et al*., 2000)
- apatite-like minerals (Uno *et al*., 2001, 2004)
- glass by low temperature vitrification (Sakuragi *et al*., 2008; Mukunoki *et al*., 2009).

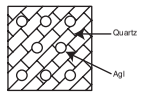
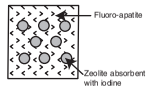
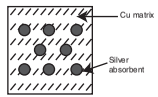
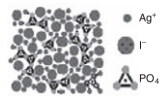
Table 5.2 summarizes several potential iodine waste forms along with their loading and anticipated performance. To date, the authors are not aware of any of these processes being utilized on an industrial scale.

5.4.3 Undissolved solids and technetium

The solid component of the irradiated fuel that does not dissolve in hot nitric acid is rich in Pd, Ru, Rh, Tc, Mo, Zr, and O. It also may contain significant concentrations of Te. The noble metals, Mo and Tc, are often in the form of the epsilon metal phase (or 5-metal phase). Molybdenum, Zr, Tc, and O are often found to vary significantly in the undissolved solids (UDS) depending on the dissolution process parameters if voloxidation is performed.

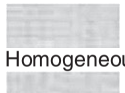
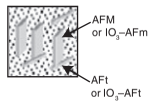
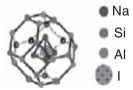
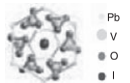
Roughly half of the technetium (depending on parameters) is dissolved. The dissolved fraction may follow uranium or the HLW raffinate depending on process parameters. The primary radionuclide to immobilize in the UDS

Table 5.2 Summary of iodine waste forms

Iodine immobilization techniques		Chemical composition		Conceptual diagrams of iodine confinement/ immobilization	Iodine release parameter
		Raw material	Waste form		
Crystalline. matrix	Rock	Spent silver-sorbent (= silica gel, AgI)	SiO_2(quartz), AgI (I content: 14 wt%)		Diffusion coefficient: 1.0 × 10^{-20} (m^2/s)
	Apatite	Fluor-apatite ($Ca_{10}(PO_4)_6F_2$): Zeolite (I sorption) = 85 : 15 (weight ratio)	Fluor-apatite ($Ca_{10}(PO_4)_6F_2$) : Zeolite (I sorption) (I content: 2 wt%)		–
	Copper matrix	Cu powder: Spent silver-sorbent = 50 : 50 (vol%)	Cu powder: Spent silver-sorbent (I content: 0.7 wt%)		Leach rate: 90 × 10^8 (Bq/y)
Glass	AgI glass	AgI : $Ag_4P_2O_7$ = 3 : 1(mol ratio)	3AgI-2Ag_2O-P_2O_5 glass (I content: 30 wt%)		Leach flux: 1.0 × 10^8 – 10^{-7} (g/ cm^2/d)

Continued

Table 5.2 Continued

Iodine immobilization techniques		Chemical composition		Conceptual diagrams of iodine confinement/ immobilization	Iodine release parameter
		Raw material	Waste form		
	BPI glass	• Glass flit : BPI = 91 : 9 (wt%) • Glass flit PbO : B_2O_3 : ZnO = 65 : 30 : 5 (mol%) • BPI : $BiPbO_2I$	6.5PbO-3B_2O_3-0.5ZnO Glass, $BiPbO_2I$ (I content: 2 wt%)		–
Cement	Hardened cememt	• Alumina cement/ $CaSO_4 2H_2O$ =100/15.5 • $NaIO_3$ conc. 0.4 mol/dm^3 • $NaIO_3$ sol./cement = 0.56	AFm: 10 wt% AFt: 46 wt% $Al(OH)_3$:44 wt% (I content: 1.85 wt%)		Distribution coefficient: >100 ml/g
Synthetic mineral	Synthetic sodalite	$NaAlO_2$: SiO_2 : NaI = 3 : 3 : 1 (mol ratio)	$Na_8(AlSiO_4)_6I_2$ (I content: 11 wt%) (measured value)		–
	Synthetic lead apatite	PbO : V_2O_5 : PbI_2 = 9 : 3 : 1 (mol ratio)	$Pb_{10}(VO_4)_6I_2$ (I content 7.2 wt%) (measured value)		–

Source: JAEA, 2007.

is ^{99}Tc and some transuranic (TRU) elements. With a half-life of 0.21×10^6 years, this waste must be immobilized for hundreds of thousands of years. In an oxidizing environment, Tc is most often found as pertechnetate (TcO_4^-), which interacts weakly with minerals and typically migrates freely in the subsurface. Two primary options are considered for these Tc-bearing wastes: (1) combine with the HLW raffinate for immobilization (e.g., in glass) or (2) form a separate waste form specifically aimed at immobilizing Tc. In the first option the noble metals (Pd, Ru, Rh) may significantly limit the loading of HLW in glass. However, addition to the HLW stream reduces the number of processes required to treat the waste and number of waste forms with very long-lived radionuclides requiring qualification.

Many waste forms have been developed for immobilization of these Tc-bearing materials:

1. An iron-based alloy that contains the UDS, reduced soluble fraction of the technetium, and potentially the soluble fraction of the noble metals and stainless steel fuel assembly hardware. This waste form strives to maintain Tc in the immobile metallic state (Tc^0). Relatively low processing temperatures (<1600°C) are required to form this alloy (Ebert, 2005).
2. The epsilon-metal phase, which makes up a large fraction of the undissolved solids, would make an outstanding waste form. This waste form incorporates the UDS, the soluble fraction of Tc, and the soluble fractions of the noble metals (Strachan *et al.*, 2010b). There are natural analogs that attest to the durability of this phase. Natural reactors operating in Gabon, Africa produced epsilon metals. The decayed Tc-99 was found to migrate less than one meter in roughly 2 billion years (Utsunomiya and Ewing 2006). That fact combined with the fact that they survive the boiling nitric acid solution despite the very high surface area attest to the high resistance to strongly oxidizing environments, despite the more extreme environment than any repository.
3. The third waste form developed for immobilization of UDS, soluble Tc, soluble noble metals, and potentially lanthanide fission products in oxide ceramics such as the mixed pyrochlore $(Zr/Nb/Mo/Tc/Ru/Rh/Pd)_2Ln_2O_7$ (Hartmann *et al.*, 2011), magnesium titanate (den Exter *et al.*, 2006), phosphates (Singh *et al.*, 2006), iron oxides/oxyhydroxides (Um *et al.*, 2011), etc. These waste forms immobilize Tc in the immobile Tc^{4+} state and display relatively high chemical durability that is independent of disposal environment.
4. Alkali-alumino silicates generated by a fluidized bed steam reforming process was studied extensively for the immobilization of Tc and I bearing wastes at Hanford (Jantzen 2008).

However, in current practice, the Tc is often vitrified with the HLW raffinate to form a borosilicate glass (e.g., at LaHague, Rokkasho, and currently planned for Hanford).

5.4.4 High-level waste raffinate

Vitrification is the process of choice for separated highly radioactive wastes in virtually every reprocessing nation. (Donald *et al.*, 1997; Ojovan and Lee, 2005; Vienna, 2005; Donald, 2010) Vitrification is:

- a proven process,
- tolerant to a wide range of waste compositions,
- a fast continuous process,
- generates no fine particulates, and
- the US Environmental Protection Agency (EPA, 2009) best demonstrated available technology (BDAT). Vitrification produces a waste form of good performance that is well understood (including many natural and ancient man-made analogs).

While vitrification into a borosilicate glass is the reference process, the next generation waste forms for HLW with potential benefits over vitrification are being developed. For example, glass composite materials (GCMs) including glass ceramics may allow for significantly higher waste loading than possible in typical borosilicate glasses (Ojovan and Lee, 2011). There are three primary limitations to the loading of HLW in glass: (i) decay heat, (ii) solubility of waste components (e.g., MoO_3), and (iii) noble metals. GCMs could allow for higher heat as the crystalline portions may be much more thermally stable. They also are expected to tolerate significantly higher concentrations of components that are sparsely soluble in the glass melt. The noble metals limit would depend on the processing methods, but, will not be more restrictive for GCMs. Crum *et al.* (2012) developed durable, radiation-resistant glass ceramics that could be processed using existing melter technologies containing roughly double the waste loading of typical glasses. Figure 5.6 shows graphically the potential for the increases in waste loading.

Other potential waste forms for HLW are crystalline ceramic waste forms, which show promise for high loading and high chemical durability (Burakov *et al.*, 2010). Development of the synthetic rock (Synroc) types of waste forms began in the 1950s; the term Synroc was coined by Ringwood *et al.* in 1979 when the most concerted waste form development and testing on these forms began (Ringwood *et al.*, 1979). Ranges of silicate, aluminate, and phosphate ceramics were developed in the 1960 to 1980s. Excellent reviews of these waste forms already exist (Lutze and Ewing, 1988; Donald, 2010; Burakov *et al.*, 2010). A number of recent advancements in these materials have shown that complicated processes such as alkoxide

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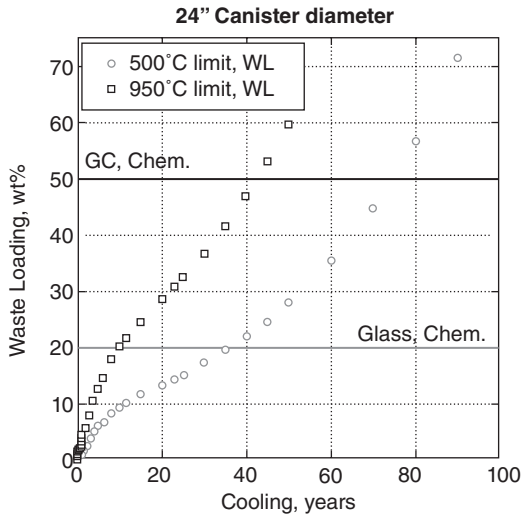

5.6 Comparison of estimated HLW raffinate loading in glass and glass ceramic waste forms. GC, Chem and Glass, Chem represent the chemical limits for glass ceramics and glass (based on data from Crum *et al.,* 2010).

precipitation and hot isostatic pressing could be replaced with a simpler melt-cast-type process (Vance *et al*., 1996; Advocat *et al*., 1997; Stefanosky *et al*., 2009).

With the advanced separations methods currently available, it is possible to subdivide the HLW raffinate into streams with similar chemical properties such as lanthanides, alkali and alkaline-earths, transition metal fission products, etc. Each of these streams could be separately immobilized in a form specifically design for the waste chemistry and disposal environment. A cost-benefit analysis was performed to evaluate the value of separating the HLW raffinate into constituent streams (Gombert *et al*., 2009). It was concluded that, aside from the noble metals, there was not a strong cost driver to further segregate the HLW. In the case of noble metals, there was a case for treating them separately under some circumstances. Thus, any further separations would be implemented for reasons other than cost.

5.4.5 Secondary waste streams

Although it is beyond the scope of this chapter to describe the quantities, treatment, and disposal of secondary wastes, it should be pointed out that these wastes make up the largest volume fractions and relatively insignificant radionuclide fractions of the waste produced in reprocessing. Mature technologies are employed to characterize, package, ship, and dispose of these secondary wastes worldwide.

5.4.6 Waste management summary

Recent advances in separations and immobilization sciences make reprocessing more effective than ever for reducing the impact of nuclear waste on the environment. Waste form development has been focused in two primary directions that tend to be competitive: (1) reduction in complexity and cost of waste treatment, storage, and disposal and (2) improvements in the long-term performance of waste forms containing long-lived radionuclides. These endeavors allow for choices to be made in how regulatory dose limits are met, and where and how limited resources are spent.

5.5 Conclusion

Management of irradiated fuel continues to challenge the nuclear power industry. Despite improvements in packing efficiency in fuel storage pools, continued delays in establishing geological repositories for permanent disposition have led utilities to move irradiated fuel into dry storage. The design for the Yucca Mountain repository in the US is in an advanced state, but this project has been suspended. Sweden and Finland have made substantial progress in locating sites and in designing geological repositories for disposing of irradiated fuel based on the once-through fuel cycles practiced in those countries. In countries such as France and Japan, irradiated fuels are processed to recycle the uranium and plutonium components for further energy production using mixed oxide fuel. Research is underway worldwide to develop advanced fuel cycle concepts that not only recycle the uranium and plutonium, but also the minor actinide component of the fuel. The goal of U/Pu recycle is to extend the supply of fuel; the primary goal of minor actinide recycle is to reduce the long-term radiological hazards associated with irradiated fuels from millions of years to a few hundred years. Regardless of the fuel cycle implemented, the choice of waste form is of critical importance to the safe disposition of the radioactive components. In reprocessing operations, consideration must be given to the waste forms used to immobilize volatile fission products, cladding, hulls, and other hardware components, undissolved solids, and the HLW stream.

5.6 Sources of further information

Yucca Mountain Repository License Application, http://www.nrc.gov/waste/hlw-disposal/yucca-lic-app.html

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6

Radioactive waste (RAW) conditioning, immobilization, and encapsulation processes and technologies: overview and advances

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Abstract: The main immobilization technologies that have been demonstrated for radioactive waste disposal are cementation, bituminization, and vitrification. Vitrification is currently the most widely used technology for the treatment of high level radioactive wastes (HLW) throughout the world. Nations that have generated HLW are immobilizing in either alkali borosilicate glass or alkali aluminophosphate glass. The compositions of nuclear waste glasses are tailored for easy preparation and melting, avoidance of glass-in-glass phase separation, avoidance of uncontrolled crystallization, and acceptable chemical durability. Future waste generation is driven by interest in sources of clean energy. The development of advanced waste forms is a necessary component of the new nuclear power plant (NPP) flowsheets. A brief summary is given of existing and advanced waste forms and processing technologies.

Key words: vitrification, cementation, bituminization, glass, cement, bitumin, waste form, advanced waste forms, conditioning, immobilization, encapsulation.

6.1 Introduction

6.1.1 Legacy waste

Most nuclear nations have generated high level radioactive waste (HLW) from nuclear weapons programs and/or commercial nuclear power generation and most store waste materials from a variety of reprocessing flowsheets (see Chapters 10–24 in this book). The Plutonium and URanium EXtraction (PUREX) process[1] is the baseline for spent fuel reprocessing

[1]The PUREX process was developed in the United States in 1950 and the world's first operational full-scale PUREX separation plant began radioactive operations at the Savannah River Plant (SRP) in 1954. The process has run continuously at SRP since start-up for defense materials only.

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for most countries with active fuel cycle programs. France and the UK reprocess spent fuel for electric utilities from other countries using the PUREX process to recover uranium (^{235}U) and plutonium (^{239}Pu). Slight modifications to the PUREX process can be made to recover ^{235}U, ^{239}Pu, ^{237}Np, and ^{99}Tc (if desired) and a number of countries (e.g., France, Japan, China, etc.) are developing solvent extraction processes to recover the minor actinides (Am and Cm) from spent fuel. Elimination of these actinides and fission products from the HLW reduces the long-term radiotoxicity and heat generation from an immobilized waste form once it is entombed in a geological repository.

Most high level waste is in one of two forms: either used nuclear fuel that is destined for direct disposal, or waste from the reprocessing of commercially generated spent nuclear fuel (SNF or commercial wastes) or from the reprocessing of fuel used to generate ^{239}Pu for weapons (defense wastes). The SNF retains a high inventory of transuranium elements (~1 at%) in its uranium matrix, and the waste from reprocessing is depleted in actinides, mainly ^{235}U and ^{239}Pu (~99% removed), having been recovered during chemical processing.

Liquid HLW streams are stored either as neutralized nitric acid streams in mild steel tanks (US and Russia) or as nitric acid streams in stainless steel tanks (France, UK, Japan, Russia). Although borosilicate glasses have become the preferred waste form for the immobilization of HLW solutions in the majority of the nuclear nations, the chemical variability of the wastes from the different reactor and reprocessing flowsheets coupled with the additional variability imposed by neutralization vs. direct storage or processing of acidic wastes has led to a diverse HLW chemistry, e.g. HLW contains about three-quarters of the elements in the periodic table.

Vitrification is currently the most widely used technology for the treatment of HLW throughout the world (Table 6.1). In the United States, more than 3,496 canisters of borosilicate glass contain vitrified, high-level waste from the Savannah River Site (defense waste processing facility) and 250 canisters at West Valley, New York. In France, approximately 14,000 canisters of HLW glass have been produced at the La Hague facility (Table 6.1).

A variety of other radioactive wastes have been generated during the fuel rod cladding/decladding processes, during chemical separations, from radioactive sources, radioactive mill tailings, medical research applications and other commercial processes such as radium for watches and clocks. Many of the sources of radioactive waste (RAW) generation are captured in other chapters in this book regarding the individual practices in various countries (includes legacy waste, currently generated waste, and anticipated future waste).

In countries where the HLW waste is neutralized before processing, the HLW has segregated into a low activity waste (LAW) fraction which is an

Table 6.1 Data on HLW glass production

Vitrification plant	Location	Melting process	Waste glass produced (metric tons)	Waste loading range (wt%)	Size of canisters (meters)	Number of canisters	TBq‡
Defense Waste Processing Facility (DWPF), Savannah River Site	Aiken, South Carolina, USA	JHCM	6,169*	28–40[c]	0.6 × 3	3,496	1.7×10^6
West Valley Demonstration Project (WVDP)	West Valley, New York, USA	JHCM	~500**	~20.4–23.5[b]	0.6 × 3	275	8.9×10^5
Waste Vitrification Plant (WVP), BNFL	Sellafield, UK	Induction, hot crucible	~~2,200[†]	~25–32[a]	0.43 × 1.34	5,627[††]	2.4×10^7
Areva NC (R7/T7)[d]	La Hague, France	Induction, hot crucible	6,642[f]	12–18[§§]	0.43 × 1	16,334	2.51×10^8
AVM or Atelier de Vitrification de Marcoule[d]	Marcoule, France	Induction, hot crucible	1,138[ξ]	12–18[§§]	0.43 × 1	3,159	1.69×10^6
Pamela	Mol, Belgium	JHCM	500[§]	15–25[§§]	0.30 × 1.2 0.43 × 1.34	2,200	4.5×10^5
Tokai Vitrification Facility[e] (TVF)	Japan	JHCM	>100	20–30[§§]	0.43 × 1	247[ff]	1.5×10^4
Mayak Vitrification Facility[f] (EP-500)	Ural Region, Russia	JHCM	~8,000	33[§§]	0.57 × 1	17,600	3.33×10^7

‡ 1 Tera-Becquerel (TBq) = 10^{12} atoms decaying per second or transmutations per second.
* 1996–June 2012.
** 1996–2002 – mission complete.
† 1991 to April 2012 at 142L glass per canister and an assumed glass density of 2.75g/cc (390kg glass per container).
†† Maximum total is 10,000 (capacity of vitrified product store), of which ~2,200 will be returned to overseas customers. Actual total is expected to be less depending on post-operation clean-out strategy.
[f] 1989–2011.
ξ 1978–2008.
§ 1985–1991.
ff 1995–2012.
§§ acidic waste loadings comprise fission products and minor actinides; corrosion products and alkali are not included as for neutralized wastes.
[a] From [163].
[b] From [204].
[c] From [205].
[d] Caterine Veyer of AREVA, personal communication (2012).
[e] Seiichiro Mitsui of JAEA, personal communication (2010).
[f] P.P. Poluektor, personal communication (2010).

alkali-rich supernate and a viscous HLW sludge fraction over time. The LAW fraction of HLW and other medium and low level wastes (MLW and LLW) can be immobilized into a variety of waste forms, i.e. cements, Ceramicrete, glass, hydroceramics, high temperature ceramic/mineral waste forms (made by a variety of technologies discussed below), glass-ceramics, and geopolymers and land disposed in safe and specially engineered facilities.

The concept of conditioning waste in order to immobilize it in solid nuclear waste forms is over 60 years old [1]. Waste forms can chemically incorporate waste species (glass, glass composite materials (GCMs), crystalline ceramics or mineral analogs, and metals), encapsulate waste species in a matrix (cement, geopolymers, hydroceramics, bitumen), or be a combination of both. Waste forms can be amorphous (glass, bitumen, geopolymers), or crystalline (crystalline ceramics including minerals and zeolites, metals, cements, hydroceramics), or a combination of forms (glass ceramic materials, GCMs; glass beads in a metal matrix; granular crystalline mineral waste-forms in a geopolymer or cement). In particular, GCMs can be formed by controlled cooling, melting above the solubility of certain waste constituents and letting them crystallize out on cooling, or by allowing homogeneous glasses at the melt temperature to cool naturally where some portion of the cooled glass crystallizes.

6.1.2 Future waste

At the turn of the twenty-first century, the interest in sources of clean energy has led to increased interest in advanced nuclear power production, often referred to as the 'nuclear renaissance.' The development of advanced waste forms is a necessary component of this new strategy. Therefore, advanced nuclear waste forms are being designed for robust disposal strategies. Implicit in the ceramics and glass ceramic waste form development is the idea of using additives to 'tailor' the waste chemically so that the desired host radionuclide phases are produced after consolidation. Implicit in the cements and encapsulant waste forms is to 'design' the encapsulant to retain radioactive and hazardous constituents while being effective in adverse environments.

6.1.3 Overview of waste forms and conditioning technologies

A brief summary of glass, glass-ceramic, crystalline ceramic (mineral), cementitious, geopolymer, bitumen, and other encapsulant waste forms is given below and in Chapter 1. There have been many comprehensive reviews of waste forms and their properties [2–11] and this will not be

elaborated on in this chapter except as examples of the different classes of waste forms discussed in Section 6.4.

This chapter will focus on the various technologies available to create the various types of waste forms and provide a cross-reference between the various processing technologies and each waste form. For example, a glass waste form can be made by Joule heated melting (JHM), advanced Joule heated melting (AJHM), plasma hearth melting (PHM), Hot Isostatic Pressing (HIP), microwave heating, and hot and cold crucible induction melting (CCIM). Thus processing technologies will be related to conditioning technologies that immobilize radioactive species either by atomic bonding or by encapsulation or a combination of the two (composite waste forms). The waste form and technology data is presented in tabulated form for brevity.

6.2 Waste form definitions

For consistency, the definitions given in Chapter 4 of this book, which are from the IAEA [12, 13], are adopted here except for the definitions of encapsulation vs. embedding where examples have been used to make the distinctions clearer.

6.2.1 Conditioning

Conditioning includes those operations that produce a waste package suitable for handling, transport, storage, and/or disposal. This may include the following:

- conversion of the waste to a solid waste form
- enclosure of the waste or waste form in containers
- providing an overpack if necessary.

The waste form is the waste in its physical and chemical form after treatment and/or immobilization prior to packaging. Thus the waste form is a component of the waste package.

6.2.2 Immobilization

Waste immobilization is the conversion of a waste into a waste form by solidification, embedding, or encapsulation. The waste form can be produced by chemical incorporation of the waste species into the structure of a suitable matrix (typically a glass, GCM, or ceramic) so that the radioactive species are atomically bound in the structure (chemical or atomic incorporation) or encapsulated.

Chemical incorporation is typical for HLW. Cementation or other encapsulation/embedding technologies are typical for LLW or ILW.

Immobilization reduces the potential for migration or dispersion of radionuclides during handling, transport, storage, and/or disposal.

6.2.3 Encapsulation and embedding

Encapsulation of waste, on the other hand, is achieved by physically surrounding the waste with or in a material (typically considered a flowable material such as a grout or cement) so the waste particles are isolated and radionuclides are retained. Encapsulation may or may not include *some chemical incorporation* if the encapsulating phase reacts with the waste, i.e. if hydrating calcium-silicate-hydrates (CSH) incorporate waste species during hydration. The IAEA definition [12] of encapsulation is 'immobilization of dispersed solids (e.g., ash or powder) by mixing with a matrix material in order to produce a waste form,' but also includes emplacement of a solid waste form (e.g., spent fuel assemblies) in a container.

Embedding is the immobilization of solid waste (e.g., metallic materials) by surrounding it with a matrix material in order to produce a waste form. Embedding is similar to encapsulation when *no chemical interaction* is observed between the waste and the encapsulation medium (typically bitumen or polymers).

6.2.4 Waste form

The IAEA defines a waste form as 'Waste in its physical and chemical form after treatment and/or conditioning (resulting in a solid product) prior to packaging.' The waste form is a component of the waste package.

A wide range of materials are potentially suitable for immobilizing RAW in a waste form. For simplicity of describing the types of waste forms in terms of chemical or atomic incorporation vs. encapsulation/embedding and the various technologies by which these waste form materials can be made, the waste forms have been grouped into ten classes:

- single-phase (homogeneous) glasses
- multi-phase glass composite materials (GCMs; heterogeneous glasses)
- single-phase crystalline ceramic/mineral analogs
- multi-phase crystalline ceramic/mineral assemblages
- bitumen
- metals
- cements
- geopolymers (inorganic) and organic polymers
- hydroceramics
- ceramicretes.

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Some waste forms are considered composites as they both atomically incorporate radionuclides and then these radio-phases are embedded or encapsulated in a secondary matrix.

Each class of waste form will be discussed in more detail in Section 6.4. Each waste class will have two tables associated with it. One table discusses the manner in which the radionuclides are sequestered (including a schematic), advantages and disadvantages of the wasteform, and the variety of immobilization technologies by which the waste form can be made. The second table provides examples of the waste form, the technology by which it is made, and references.

6.3 Types of immobilization processes and pre-processes

Various thermal and non-thermal processes can be used to make various types of waste forms. The cross-referencing of the various processing technologies that can be used to produce various waste forms is given in Table 6.2. In Table 6.2 the technologies are designated as continuous processes which are more applicable to large volume wastes, or batch processes which are more applicable to small to medium volume wastes. Table 6.2 provides a crosswalk of the processes/technologies (rows) that can be used to form various waste forms (columns). A list of advantages, disadvantages, and types of waste form that can be made with a particular technology are also given in Table 6.2. Further discussion of the individual technologies (rows) can be found in Chapter 4 of Reference 11.

The processing and pre-processing technologies by which a waste form can be made are briefly described below as thermal or non-thermal technologies in keeping with Table 6.2.

Thermal processes

- Calcination – heating at elevated temperature to convert all cations to the oxide form (removes waters of hydration, hydroxides, nitrates in the presence or absence of air, i.e. rotary pyrolytic calciners). May be coupled with other high temperature processes.
- Drying – heating at 110°C to remove bound water in preparation for solidification, embedding or other high temperature processes.
- Vitrification – the process of solidifying a liquid, sludge, solid, thermal residue, granular waste form, or calcine in a glass (borosilicate, iron phosphate, aluminosilicate).
- Metal formation – melting a metallic waste with or without other metal additives.

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Table 6.2 Waste form processing technologies

	Processing technology	Process mode	Treatment and waste stream scale	Waste forms produced	Advantages	Disadvantages
Thermal technologies	Joule Heated Melter (JHM)	Continuous	Large	Borosilicate glass, other glasses (LaB's, FeP, AlP, chalcognide, etc.)	Proven technology; typically operates with a 'cold cap' to minimize volatility of species of concern	Electrode and refractory erosion may be a problem; solubility control of certain species (Cr, Mo, and SO_4) critical
	Advanced Joule Heater Melter (AJHM)	Continuous	Large	Borosilicate glass, GCM's, other glasses (LaB's, FeP, AlP, chalcognide, etc)	Increased capacity, throughput, and melt rate compared to JHM	Operates with minimal or no 'cold cap' with associated increases in volatility of species of concern
	Cold Crucible Induction Melter (CCIM)	Continuous	Large	Borosilicate glass, GCM's, other glasses (LaB's, FeP, AlP, chalcognide etc), crystalline ceramics, simple oxides, metal matrix	Allows processing of corrosive glasses; no refractories; no electrodes; water cooled; can be stirred if needed; increased capacity compared to JHM and AJHM; can operate at higher temperatures than JHM and AJHM; operates with a 'cold cap' to minimize volatility	Higher temperature operation can increase volatilization of species of concern but 'cold cap' coverage minimizes these impacts

Thermal technologies	In-Container Vitrification (ICV); also known as 'Bulk Vitrification'	Batch	Depends on container size (could be medium to large)	Borosilicate glass; GCM's, Other Glasses (LaB's, FeP, AlP, chalcognide etc.)	Inexpensive and simple for low activity wastes or contaminated soils; not applicable to HLW	Inhomogeneous waste forms produced; no temperature control so radionuclide vaporization is high; little or no convection in melt
	Self-Sustaining Vitrification (SSV)	Batch	Small	GCM's	Inexpensive; can be used to process small amounts of wastes at remote locations	May require some pre-processing, i.e. grinding of the waste and pre-mixing
	Cold Press and Sinter (Cold Uniaxial Pressing, CUP: Cold Isostatic Pressing, CIP)	Batch	Small	GCM's, crystalline ceramics, simple oxides, metal matrix, zeolites, hydroceramic	Higher waste loadings; Minimum disposal volume	Usually small scale; may require pre-calcining or pre-treating waste to an oxide to avoid shrinkage of form
	Hot Isostatic Pressing (HIP)	Batch	Small	Borosilicate glass (lab scale only), GCM's, crystalline ceramic/simple oxides, metal matrix, zeolites, hydroceramics	Zero off-gas emissions; higher waste loadings; minimum disposal volume; mature flexible technology; no major secondary wastes; mature industrial process	Processes small quantities; can overpressurize if large amounts of volatiles (e.g. nitrates/ hydrates) are present; may require pre-calcining or pre-treating waste to an oxide (shrinkage handled by bellows like canisters)

Continued

Table 6.2 Continued

	Processing technology	Process mode	Treatment and waste stream scale	Waste forms produced	Advantages	Disadvantages
Thermal technologies	Hot Uniaxial Press (HUP)	Batch	Small	Borosilicate glass (lab scale only); GCM's, crystalline ceramic, simple oxides, metal matrix, zeolites, hydroceramic	Higher waste loadings; minimum disposal volume, mature flexible technology; mature industrial process	Usually small scale; may require pre-calcining or pre-treating waste to an oxide for shrinkage control
	Cyclone Furnaces	Continuous	Large	Borosilicate glass, GCM's, other glasses (LaB's, FeP, AIP, chalcognide, etc.), crystalline ceramics, simple oxides, metal matrix	Suitable for soils containing low volatility radionuclides	Secondary recovery process needed to treat off gases

	Fluidized Bed Steam Reforming (FBSR)	Continuous	Large	Crystalline ceramic, simple oxides, zeolites	Pyrolysis (not incineration); immobilizes halides, sulfates, ^{99}Tc sequestered in sodalite; moderate temperature; ≥85% volatile species contained; wastes processed without neutralization; destroys organics and nitrates; industrially proven technology; no secondary liquid waste stream	Product is granular and requires a high integrity container (HIC) or encapsulation in a binder to make a glass ceramic material, a geopolymer, or a hydroceramic; Radionuclide partitioning amongst the phases needs to be further studied
Thermal technologies	Electric Arc Furnaces	Batch	Medium/Large	High temperature glasses, GCM's, crystalline ceramics, simple oxides, metal matrix	Established Industrial Practice; Similar technology is used for ICV	No large-scale radioactive practice; high temperatures; volatilization of radionuclides
	Plasma Furnaces	Batch	Small	Borosilicate glass, high temperature glasses, GCM's, crystalline ceramics, simple oxides, metal matrix	Plasma generating electrode erosion; efficient for the destruction of organics	Large-scale practice in Belgium (Belgoprocess; high temperatures; volatilization of radionuclides [206–208])

Continued

Table 6.2 Continued

	Processing technology	Process mode	Treatment and waste stream scale	Waste forms produced	Advantages	Disadvantages
	Microwave Heating	Batch	Small	Borosilicate glass, GCM's, other glasses (LaB's, FeP, AIP, chalcognide, etc.), crystalline ceramics, simple oxides, metal matrix	Suitable for mixed wastes; Can be used as a heat source in other equipment (e.g fluidized bed)	Limited to small scale; process scale up; inhomogeneous heating (need a susceptor material); no large-scale practice
Non-thermal technologies	Cement	Continuous or Batch	Large	Ordinary portland cement (OCP), High Alumina Cements, Geopolymeric Cements with Fly Ash, slag, or meta-kaolin	Simple technology; design formulation for best waste retention; fly ash and slag additives keep ^{99}Tc and Cr in reduced oxidation state to prevent leaching	Formulations waste specific; some sequestering of radionuclides in hydration products vs. grain boundaries needs more study; radiolytic production of H_2 in high radiation; pH of pore water alkaline and promotes leaching

Non-thermal technologies	Geopolymer	Batch	Large or Small	Geopolymers incorporate liquid waste encapsulate incinerated, pyrolyzed, or calcined wastes, geopolymeric cements	Minimal water so radiolytic H_2 generation is limited, fire resistant, pore water less alkaline than cements	Formulations waste specific; distribution of radionuclides among the phases needs more study; batches are thick and require extrusion
	Hydroceramics	Batch	Small	Zeolite, crystalline ceramic	High capacity for high sodium or calcium containing wastes; stabilize halides and sulfates.	Require hydrothermal set; requires more water than geopolymers so radiolytic H_2 generation; batches are thick and require extrusion; wastes with >25wt% nitrate must be pre-treated
	Ceramicrete	Batch	Small	Crystalline ceramic incorporates liquid waste or encapsulates	Very dense; room temperature curing; high waste loading	High heat of hydration; bubble formation which can be vibrated out of mixture during set
	Bitumen	Continuous or batch	Large to small	Encapsulated or embedded waste forms	Simple; low operating cost; leach-resistant characteristics	Flammable; requires heat to make bitumen molten; poor performance with salts; thick even when molten; requires extrusion

Adapted from [11].

- Pyrolysis – process of destroying organics in the absence of air (more environmentally compliant than incineration which destroys organics in the presence of air). Pyrolysis can be carried out in calciners, drums, or by fluidized bed steam reforming (FBSR).
- Hot isostatic pressing (HIP) – a manufacturing process used to reduce the porosity of metals and increase the density of many ceramic materials by subjecting the waste/additive mixture to both elevated temperature and isostatic gas pressure in a high pressure containment vessel.
- Cold isostatic pressing (CIP) and sintering – a manufacturing process used to reduce the porosity of metals and increase the density of many ceramic materials by subjecting the waste/additive mixture to isostatic liquid pressure in a flexible but impervious form such as a balloon before sintering at high temperature.
- Hot uniaxial pressing (HUP) – a manufacturing process used to reduce the porosity of metals and increase the density of many ceramic materials by subjecting the waste/additive mixture to uniaxial mechanical pressure from above and below in containment form while simultaneously subjecting the form to elevated temperature.
- Cold uniaxial pressing (CUP) and sintering – a manufacturing process used to reduce the porosity of metals and increase the density of many ceramic materials by subjecting the waste/additive mixture to uniaxial mechanical pressure from above and below in containment form before sintering at high temperature either with or without the containment form.

Non-thermal processes

- Cementation – the process of solidifying a liquid, sludge, solid, thermal residue, granular waste form, or calcine in cement matrix of crystalline calcium silicates, aluminates, and ferrate.
- Geopolymerization – the process of solidifying a liquid, sludge, solid, thermal residue, granular waste form, or calcine in an amorphous sodium aluminosilicate matrix.
- Bituminization – the process of solidifying a liquid, sludge, solid, thermal residue, granular waste form, calcine in bitumen.
- Forming – mixing a waste with cementitious, geopolymeric, bituminous, hydroceramic, or Ceramicrete-type additives and mixing in a form, i.e. can, vault, canister, and allowing the material to set or age.
- Pouring – similar to forming but the waste/additive mixture can be poured, extruded, or emptied into a form to set or age.
- Compositing – using metals, glass, cements, geopolymers, etc, to encapsulate a waste that has already been solidified for special reasons such

as heat dissipation, control of respirable fines in calcined or granular waste forms, and/or compressive strength requirements.

Often processes are coupled. For example, in France and the UK waste is calcined to remove excess nitrates before vitrification into a final waste form. This allows free-flowing oxides to enter the melter without nitrates being off-gassed or causing the particles to adhere to one another. Organic bearing wastes are often pyrolyzed to remove organics, if needed, before vitrification [14, 15].

Calcining is often performed before HIP, CIP, HUP, or CUP processes are performed, so that volatile species are not given off during the hot pressing or during the subsequent sintering. This ensures that the pressed waste form retains its integrity and form and does not crack during processing from off-gassing of hydrated or nitrated species.

6.4 Immobilization processes and technologies

The major types of waste forms will be described in regard to the manner in which the radionuclides are immobilized and the methods by which each can be made. Different waste forms give different durability tests responses. Single-phase waste forms (glass and single-phase oxides or crystalline ceramics (minerals) have only one source of radionuclides that can leach during a durability test. In multiphase waste forms the distribution of the radionuclides amongst the phases present becomes important as each phase has its own rate of leaching for the specific elements that it sequesters. Each waste form given in Tables 6.3–6.10 will be described in terms of the radionuclide immobilization achieved and references given as to which conditioning technologies can be used to make each type of wasteform.

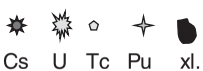
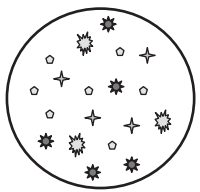
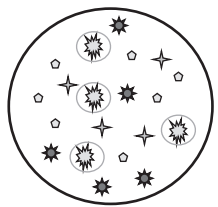
The immobilization of HLW is always achieved by its *atomic-scale* incorporation into the structure of a suitable matrix (typically glass, a GCM, or a crystalline ceramic (also sometimes referred to as mineral analog waste forms) so that the radionuclides are incorporated into durable structures by any combination of short range order (SRO),[2] medium range order (MRO)[3] or long range order (LRO).[4] Glasses incorporate radionuclides and hazardous species into their atomic structure by SRO and MRO [16]. Recent experimentation has shown the existence of large cation-rich clusters in glass, e.g. clusters of Ca in $CaSiO_3$ glasses and clusters of Na_2MoO_4

[2]SRO: radius of influence ~1.6–3 Å around a central atom, e.g. polyhedra such as tetrahedral and octahedral structural units.

[3]MRO: radius of influence ~3–6 Å encompasses second- and third-neighbor environments around a central atom. The more highly ordered regions, referred to as clusters or quasicrystals, often have atomic arrangements that approach those of crystals.

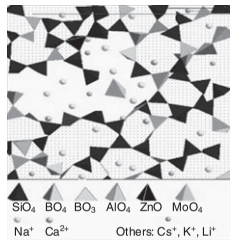
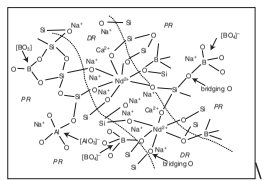
[4]LRO extends beyond third-neighbor environments and gives crystalline ceramic/mineral structures their crystallographic periodicity.

Table 6.3 Attributes of homogeneous vs. inhomogeneous glass waste forms

Waste form	Homogeneous glass	Inhomogeneous glass
Description	Radionuclides and hazardous species are atomically bonded in a durable glass structure usually to oxygen atoms that are also bonded to the matrix elements, Si, Al, B, P, etc., by short range order (SRO) and medium range order (MRO)	Some radionuclides and hazardous species are atomically bonded in a durable glass structure as with homogeneous glass, but other radionuclides reside in a very soluble immiscible glass phase (glass-in-glass phase separation)
Radionuclide immobilization mechanism Key 	Chemical incorporation 	Chemical incorporation 
Waste loading(s)/ durability	(a) moderate waste loading, (b) good overall durability, (c) easy to model radionuclide release from a single phase	(a) moderate waste loading, (b) poor durability for certain radionuclides, (c) impossible to model radionuclide release as the fraction of the second phase is dependent on thermal history
Immobilization technologies	Joule Heated Melters (JHM), Advanced Joule Heated Melters (AJHM), Cold Crucible Induction Melters (CCIM), Hot Isostatic Pressing (HIP), Hot Uniaxial Pressing (HUP)	Joule Heated Melters (JHM), Advanced Joule Heated Melters (AJHM), Cold Crucible Induction Melters (CCIM), Hot Isostatic Pressing (HIP), Hot Uniaxial Pressing (HUP)

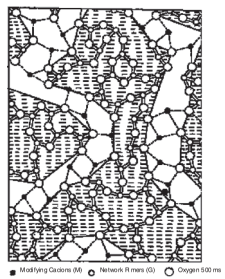
Adapted from [11].

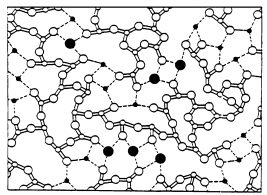
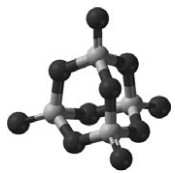
Table 6.4 Examples of homogeneous glass waste forms demonstrating their SRO and MRO structure

Type of glass	Major structural components		Comments
Alkali borosilicate [3–8, 18, 34, 61]	$(SiO_4)^{-4}$, $(BO_4)^{-5}$, $(BO_3)^{-3}$ and some $(AlO_4)^{-5}$ and $(FeO_4)^{-5}$ structural units to which alkali, alkaline earth, and waste species bond.	 Atomic structure of a French nuclear waste glass: unshaded region shows formation of a $(Na,Cs)_2MoO_4$ cluster [209].	Ease of processing, melt temperatures 1150–1200°C to minimize volatility; cold cap production if feasible minimizes volatility; most waste cations highly soluble in glass; overall waste solubility 25–40 wt%; made by JHM, AJHM, induction melting, CCIM or HIP.
Lanthanide borosilicate (LaBS) [5,6, 18, 210–214]	$(SiO_4)^{-4}$, $(BO_4)^{-5}$, $(BO_3)^{-3}$and some $(AlO_4)^{-5}$ structural units to which lanthanides, alkaline earth, and other waste species bond.	 Atomic structure of a French HLW rare earth bearing borosilicate glass. Na^+, Ca^{2+} and Nd^{3+} exist in the percolation channels. PR is the polymerized region and DR is the depolymerized region [7].	Higher waste loading (16–59 wt%) for actinides/lanthanides than alkali borosilicates; lanthanides serve as neutron absorbers; 1300–1500°C melting causes volatilization of some radio-nuclides; corrosion similar to alkali boro-silicates; made by CCIM, HIP, or induction melting.

Continued

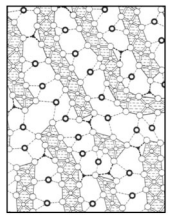
Table 6.4 Continued

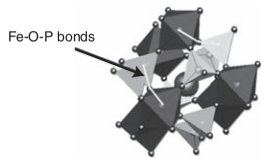
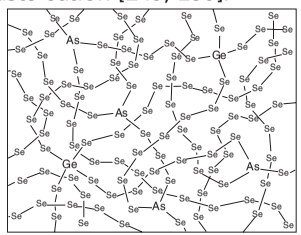
Type of glass	Major structural components		Comments
Aluminosilicate glasses and/or alkali aluminosilicate glasses [4, 8, 61]	$(SiO_4)^{-4}$ and $(AlO_4)^{-5}$ structural units to which alkali, alkaline earth, and waste species bond (similar structure to borosilicate glasses when $(BO_4)^{-5}$ are present).	 Atomic structure of a simple generic $M_2O_3(G_2O_3)_2$ glass (M is modifying cations, G represents tetrahedral cations). The shaded regions are the PR regions. The un-shaded regions represent the percolation channels or DR regions (from [215]).	Melt temperature of ~1600°C causes volatilization of radionuclides; waste loading dependent on rapid cooling, e.g. 20 wt% UO_2 if cooled rapidly while <10 wt% if cooled slowly; improved durability over borosilicate glass; CCIM, HIP.

Aluminoborate glasses	$(BO_4)^{-5}$, $(BO_3)^{-3}$ and some $(AlO_4)^{-5}$.	Not used for waste vitrification.	
High silicate glasses (sintered glasses) [4, 8, 61]	$(SiO_4)^{-4}$	 Atomic structure of sodium silicate glass. Glass formers are small open circles, oxygen atoms are large open circles, modifier cations are small filled circles, U atoms which form clusters are large filled circles [216].	Requires hot pressing and sintering at 600–800°C in order to retain volatile fission elements such as Cs, Ru, Mo and Tc; waste solubility 5–35 wt%.
Alkali alumino-phosphate [3–8, 34, 217–220]	$(PO_4)^{-3}$ and $(AlO_4)^{-5}$ structural units to which alkali, alkaline earth, and waste species bond.	 Atomic structure of phosphate glass with P_4O_{10} cage-like structures which provide the basic building block for phosphate glass formers.	Melts at lower temperatures than silicate or borosilicate systems; most cations readily incorporated; accommodates >10 wt% sulfate; corrosive to materials of construction; tendency to devitrify; durability comparable to borosilicate glass if alumina content is sufficient; composition ~ 24–27 Na_2O, 20–24, $Al_2O_3 + Me_mO_n$, 50–52 P_2O_5; JHM, AJHM, CCIM.

Continued

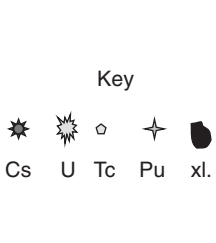
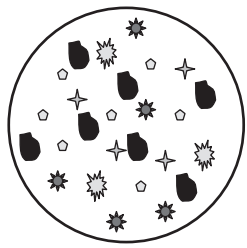
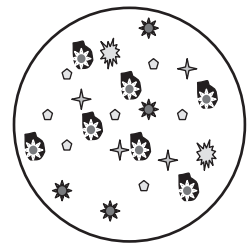
Table 6.4 Continued

Type of glass	Major structural components		Comments
Lead iron phosphate (LIP) [4, 8, 61, 221–227]	$(PO_4)^{-3}$ and $(FeO_4)^{-5}$ structural units to which alkali, alkaline earth, and waste species bond.	 Atomic structure of LIP glass. Polyphosphate chains are cross-linked by lead atoms (open circles) and iron atoms (small filled circles) which form 'knots' in the percolation pathways that inhibit cation diffusion [228].	40–66 PbO; 30–55 P_2O_5; 0–10 Fe_2O_3 dependent on amount of iron oxide in waste; melts 850–1050°C; waste loading (~20 wt%); abandoned due to regulatory issues with PbO component poor solubility of certain species, devitrification, poorer waste solubility than borosilicate glasses, etc.; JHM, AJHM, CCIM.

Iron phosphate (IP) [229–247]	$(PO_4)^{-3}$ and $(FeO_4)^{-5}$ tetrahedral structural units to which alkali, alkaline earth, and waste species bond; Fe-O-P bonds have shown to be hydration resistant whether iron is Fe^{2+} or Fe^{3+} [248]	 Atomic structure of IP glasses are nano-heterogeneous, with $FePO_4$-like regions and phosphate chains that incorporate Fe^{2+}/Fe^{3+} network-modifying cations. Large atom in center of cage like structure is a waste cation [249, 250].	Good chemical durability; high solubility for many heavy metals (U, Cr, Zr, Cs, Mo, noble metals, rare earths); melts 950–1100°C; viscosity typically <1 poise; low corrosion of oxide refractories and Inconel alloys; waste loadings 25–50 wt%; tendency to devitrify; JHM, AJHM, CCIM.
Chalcogenide and chalcohalide [96, 251, 252]	Glasses obtained by melting chalcogen elements (S, Se, and Te) with Group V and IV elementsTeO_2-*X*Cl-$B_2O_3TeO_2$-*X*Cl-Li_2OTeO_2-*X*Cl-Na_2O*X*Cl = 'mixed chlorides' waste simulant at ~19 wt%	 [from 253]	S, Se, and Te glasses for radionuclides difficult to immobilize in borosilicate glass systems, i.e. ^{129}I. Gels such as $Pt_2Ge_4S_{9.6}$ are used to immobilize actinides, noble gases, carbon dioxide, and mixed chlorides.

Adapted from [11].

Table 6.5 Attributes of glass-ceramics and glass composite material (GCM) waste forms

Waste forms	GCM (secondary crystalline phase contains no radionuclides and/or is inert)	GCM (secondary crystalline phase contains radionuclides and should be durable)
Description	Radionuclides can be chemically incorporated in the glassy matrix (same as single phase glasses) and crystals such as spinels (Cr, Ni, and Fe species) crystallize that do not contain radionuclides and are inert.	Radionuclides can be chemically incorporated in the glass matrix and in the crystalline phases. Example shows Cs in the glass and in a secondary phase. Secondary phases need to be durable like pollucite $(Cs,Na)_2Al_2Si_4O_{12}$ and soluble phases such as $(Na,Cs)_2SO_4$ should be avoided as they are not GCMs.
Radionuclide immobilization mechanism	Chemical incorporation	Chemical incorporation and encapsulation
		
		
Waste loading(s)/ durability	(a) higher waste loadings, (b) secondary phases have no radionuclides, (c) good overall durability, (d) easy to model radionuclide release from single phase glass once grain boundary dissolution is experimentally shown to be minimal	(a) higher waste loadings, (b) secondary phases contain long-lived radionuclides, (c) glassy phase can contain the shorter lived radionuclides or no radionuclides, (d) more complex to model radionuclide release from multiple phases and grain boundaries
Immobilization technologies	Joule Heated Melters (JHM – crystals form on cooling), Advanced Joule Heated Melters (AJHM ~1–3 vol% crystals probable), Cold Crucible Induction Melters (CCIM 10–50 wt% crystals), Hot Isostatic Pressing (HIPing >40 wt% crystals), Hot Uniaxial Pressing (HUPing >90 wt% crystals), Press and sinter (> 90 wt% crystals)	

Adapted from [11].

Table 6.6 Examples of glass-ceramics and glass ceramic materials (GCM) as waste forms

Name	Glass phase	Crystalline phase(s)	Comments
Borosilicate based			
Alkali borosilicates [44, 45, 254–257]	Borosilicate	$NiFe_2O_4$ spinels, $ZrSiO_4$, Al_2O_3	Glasses which are allowed to partially crystallize in a stirred melt pool or upon cooling; crystals are inert; crystallized glass viscosity is non-Newtonian; secondary phases must be inert; AJHM or CCIM.
Glass bonded sodalites [49–51, 258]	Borosilicate	^{129}I in NaI, ^{129}I in sodalite, $Na_8Al_6Si_6O_{24}(I)_2$, Cl in sodalite, $Na_8Al_6Si_6O_{24}(Cl)_2$	Electrorefiner wastes; radionuclide release from each phase is measured, e.g. Si, Al, Na, Li (sodalite and glass), B (glass), Cl, I (sodalite and halite); HIP or cold pressing/sintering.
Synroc alumino-borosilicates [11]	Alumino-borosilicate	Zirconolite, $CaZrTi_2O_7$	Zirconolite is major crystalline phase for Pu and Gd (neutron absorber); for low purity actinide wastes; Pu partitions into crystalline phase over the glass phase by a factor of 100:1; accommodates actinides and any associated impurities; HIP.
Barium aluminosilicates celsian [4, 8, 61, 259]	Borosilicate (sodium alumino-silicate with 2–7 wt% B_2O_3 and 3–4 wt% TiO_2)	Celsian, $BaAl_2Si_2O_8$ pyrochlore ($RE_2Ti_2O_7$; *RE*-rare earth), Scheelite ($BaMoO_4$), Pollucite ($CsAlSi_2O_6$) molybdenum-nosean [$Na_8Al_6MoO_4(SiO_4)_6$], Perovskite $CaTiO_3$, Diopside $CaMgSi_2O_6$, Eucryptite $LiAlSi_2O_6$ spodumene $LiAlSi_2O_6$, Nepheline, $NaA1SiO_4$	Pyrochlore host for actinides and Sr; pollucite host for Cs and Rb; noble metal fission products form small metallic droplets. Melt temperatures from 1100–1400°C; controlled crystallization between 530 and 720°C; leaching characteristics have been noted to be comparable to the borosilicate glasses affording no significant advantages; additional work in this area has been limited, melt and control crystallization or press and sinter.

Continued

Table 6.6 Continued

Name	Glass phase	Crystalline phase(s)	Comments
Diopside borosilicates [4, 8, 61, 259, 260]	Borosilicate	Diopside $CaMgSi_2O_6$, Powellite $CaMoO_4$, Perovskite $CaTiO_3$	Waste loadings ~ 30 wt% for European and Japanese commercial wastes which is usually ~16 wt%; Melted at 1300°C; controlled crystallization in the range 800–1100°C; Cs was in the diopside; La, Ce, Nd, Pr in the perovskite, Sr and Sm were in the glass; noble metals were metallic.
Titania based			
Synroc and sphene [4, 8, 61, 261–270]	Sodium aluminosilicate with TiO_2 and CaO	Sphene $CaTiSiO_5$, Pyrochlore Ca(RE,U) Ti_2O_7, Zirconolite $CaZrTi_2O_7$, Perovskite (Ca,Re,U,Sr), TiO_3, Anorthite $CaAl_2Si_2O_8$	Sphene and Synroc crystalline ceramic forms, mainly zirconolite, can also be formulated. Formation at 1300–1500°C. Actinides and REEs, and Sr are in zirconolite; Cs and the remaining Sr into the vitreous phase; CCIM and cool, press and sinter.
Alkali titanium silicate [4, 8, 61, 271]	Sodium Titanium silicates	Corundum Al_2O_3, Cristobalite SiO_2, Albite $NaAlSi_3O_8$, Zirconolite $CaZrTi_2O_7$, Perovskite (Ca,Re,U,Sr) TiO_3, Zircon $ZrSiO_4$	Formed by HIPing calcine (70 wt%) with Si, Ti, Al metal and alkali oxides; for high Zr containing Idaho National Laboratory wastes.
Barium titanium silicate Fresnoite [4, 8, 61, 261]	Barium silicates with TiO_2	Fresnoite, $Ba_2TiSi_2O_8$, Ba priderite $BaFe_2Ti_6O_{16}$, Pyrochlore $RE_2Ti_2O_7$, Scheelite $BaMoO_4$	Form at 1200°C. Fresnoite hosts Ba and Sr, priderite hosts Ba, pyrochlore hosts RE, actinides, RE and Sr. Cs remains in the glassy phase. Glass is 50% and crystalline phases are 50%.

Silicate based			
Basalt [4, 8, 61, 261. 272, 273]	Complex natural oxide based on Si, Ca, Mg, Fe, Al and Ti	For Purex wastes: augite (Ca, Mg, $Fe)_2Si_2O_6$ powellite (Ca, Sr) MoO_4 spinel ($NiFe_2O_4$).	Glasses melt in the range 1300–1400°C. Crystallization is carried out at temperature ranges 670–700°C and 900–950°C. The chemical durability superior to that of borosilicate glasses; JHM, CCIM.
Iron enriched basalt (IEB) [4, 8, 61, 261]	Alumino-silicate glass	Iron spinel; feldspars $NaAlSi_3O_8$ to $CaAl_2Si_2O_8$; augite $(Ca,Mg, Fe)_2Si_2O_6$; fluroapatite, $Ca_5(PO_4)_3F$; zircon, $ZrSiO_4$; fluorite CaF_2; cristobalite SiO_2; hematite, Fe_2O_3, mullite $Al_6Si_2O_{13}$	Applications to commercial and defense wastes, including decontamination of Three Mile Island containment water together with core debris; melt at 1400–1500°C and controlled cooling after casting the glass into containers; JHM, CCIM; arc melting.
Iron enriched basalt (IEB) with TiO_2 and ZrO_2 [4, 8, 61, 261]	Alumino-silicate glass	Same as above plus: zirconolite, pseudobrookite Fe_2TiO_5, chevkinite $Ce_4Fe_2Ti_3Si_4O_{22}$	Cast glasses crystallized by holding at 1200°C for 16 h; Ti phases retain the actinides; JHM, CCIM; arc melting.
Magnesium aluminosilicate (MAS) [4, 8]	Magnesium alumino-silicate	Enstatite $MgSiO_3$Indialite/Corderite $Mg_2Al_4Si_5O_{18}$	Used as an encapsulant for Zr alloy cladding wastes; accommodates 20% ZrO_2; press and sinter.
Phosphate based			
Apatite/monazite glass ceramics [274–276]	Calcium phosphate	Apatite $Ca_5(PO_4)_3(F,Cl)$Monazite (Ce,U) PO_4	Apatite hosts Ca,P,F, Cl, S, Sr, Cs, As, Pb, Ba, Hg, Cd, Cr, U, and Ce, melted at 1400°C, crystallized at 1150°C and allowed to furnace cool; investigated primarily for phosphate-rich or fluoride-rich waste streams including Idaho National Laboratory CaF_2 wastes; JHM, AJHM, CCIM.

Adapted from [11].

Table 6.7 Attributes of homogeneous and multiphase ceramic (mineral) waste forms

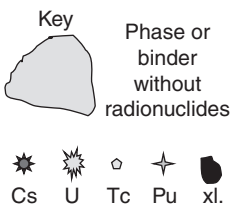
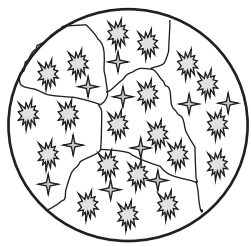
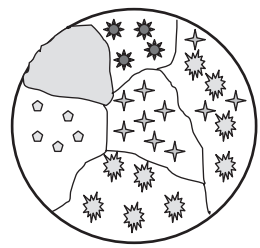
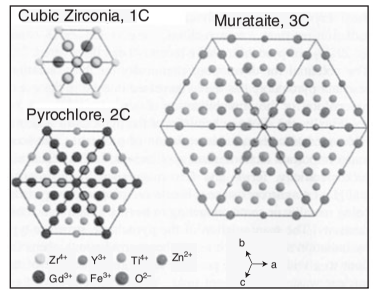
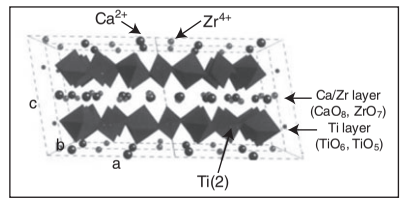
Waste form(s)	Single phase oxides/ minerals/metals (granular or monolithic)	Multiphase oxides/minerals/ metals (granular or monolithic)
Description	Individual phases contains one radionuclide or hazardous species or a solid solution, i.e. UO_2-ThO_2 (shown).	Individual phases contain different or multiple radioactive or hazardous species (see solid solution indicated between UO_2-ThO_2). Some phases do not incorporate radionuclides or hazardous species at all.
Radionuclide immobilization mechanism	Chemical incorporation	Chemical incorporation
		
Waste loading(s)/ durability	(a) high waste loading for single radionuclide or hazardous species (b) durability (c) easy to model species released from a single phase (d) may require precalcining for certain technologies to work efficiently	(a) high waste loadings (b) superior overall durability (c) difficult to model durability of species released from multiple phases and grain boundaries (d) need to tailor for species partitioning amongst phases (e) need to determine species partitioning and source terms from each phase (f) may form an intergranular glassy phase that sequesters species of concern (e) may require precalcining for certain processes to work efficiently
Immobilization technologies	Hot Isostatic Pressing (HIPing >40 wt% crystals), Hot Uniaxial Pressing (HUPing >90 wt% crystals), Press and sinter (> 90 wt% crystals), Fluidized Bed Steam Reforming (>90 wt% crystals)	

Table 6.8 Examples of single and multiple crystalline ceramic (mineral) waste forms

Crystalline ceramic phase	Comments	Structure
Simple oxides		
XO_2 Oxides [277–280]	ZrO_2, UO_2, ThO_2,HfO_2, PuO_2 have the simple fluorite CaF_2 cubic structure; make by HIP, HUP, press and sinter, melt and crystallize.	
Complex oxides		
Pyrochlore [281–284]	A derivative of the fluorite structure type, $A_2B_2O_7$, where A-site contains large cations (Na, Ca, U, Th, Y and lanthanides) and the B-site contains smaller, higher valence cations (Nb, Ta, Ti, Zr, Fe^{3+}).	
Murataite [3, 7, 285–293]	Also a derivative of the isometric fluorite structure $A_6B_{12}C_5TX_{40-x}$. with multiple units of the fluorite unit cell; hosts U, Pu, Cm, and REs including Gd a neutron absorber. Forms in solid solution with pyrochlore.	
Zirconolite [294–299]	Monoclinic $CaZrTi_2O_7$ has a fluorite-derived structure closely related to pyrochlore, where Pu, U, Gd and Hf may be accommodated on the Ca/Zr-sites, as in the case of $Ca(Zr,Pu)Ti_2O_7$.	



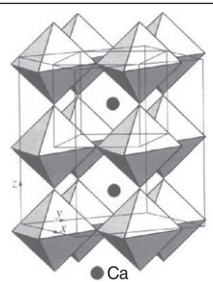
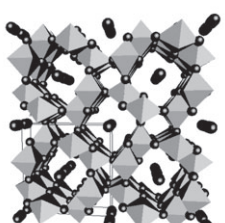
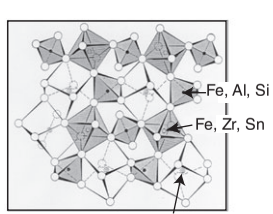
Projection along [111] direction for zirconia, pyrochlore, and murataite structures [from 284]

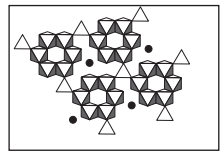
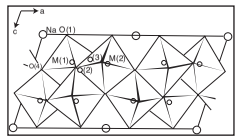
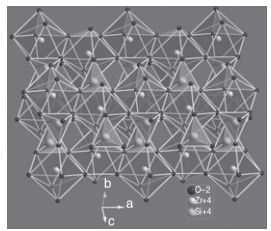


[from 7]

Continued

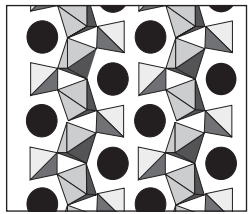
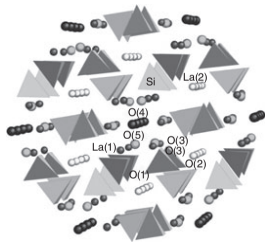
Table 6.8 Continued

Crystalline ceramic phase	Comments	Structure
Perovskite [296, 300]	$CaTiO_3$ has a wide range of compositions as stable solid-solutions; orthorhombic; consists of a three-dimensional network of corner-sharing TiO_6 octahedra, with Ca occupying the large void spaces between the octahedra (the corner-sharing octahedra are located on the eight corners of a slightly distorted cube). Plutonium, other actinides, and rare-earth elements can occupy the Ca site in the structure, as in (Ca,Pu)TiO_3. The octahedra can also tilt to accommodate larger cations in the Ca site [from 301].	 [from 301]
Ba-Hollandite [302, 303]	$Ba_{1.2}(Al,Ti)_8O_{16}$ tunnels between TiO_6 octahedra accommodate ^{133}Ba, ^{137}Cs and ^{90}Sr.	
Ferrite garnet [283]	$^{[8]}A_3{}^{[6]}B_2[TiO_4]_3$, e.g. $^{[8]}(Ca,Gd,\text{actinides})^{[6]}Fe_2{}^{[4]}Fe_3O_{12}$	
Garnet [304–307]	$A_3B_2(XO_4)_3$; distorted cubic structure; BO_6 octahedra and XO_4 tetrahedra establish a framework structure alternately sharing corners; A and B sites can host actinides, REs, and X = Si^{4+}, Fe^{3+}, Al^{3+}, Ga^{3+}, Ge^{4+} and V^{5+} making silicate, ferrite, aluminate, gallate, germinate, and vanadate garnets.	

Crichtonite [308]	$(Sr, La, Ce, Y)(Ti, Fe^{3+}, Mn, Mg, Zn, Cr, Al, Zr, Hf, U, V, Nb, Sn, Cu, Ni)_{21}O_{38}$	 Sr, La, Ce, Y positions are indicated by the solid circles. Other cations are in the octahedral positions. [from 309]
Freudenbergite [310]	$Na_2Al_2(Ti,Fe)_6O_{16}$ a spinel-based phase suitable for incorporating Al-rich wastes from Al fuel cladding/decladding. The A site can accommodate Na,K while the different octahedral sites can accommodate Mg, Co, Ni, Zn, Al, Ti^{3+}, Cr, Fe, Ga, Si and Nb.	 [from 311]
Simple silicates		
Zircon/Thorite [312, 313]	$ZrSiO_4/ThSiO_4$; zircon is an extremely durable mineral that is commonly used for U/Pb age-dating, as high uranium concentrations (up to 20,000 ppm) may be present; the $PuSiO_4$ end member is known and Ce, Hf and Gd have been found to substitute for Zr.	

Continued

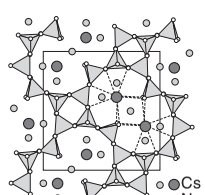
Table 6.8 Continued

Crystalline ceramic phase	Comments	Structure
Titanite (sphene) [261, 314]	$CaTiSiO_5$ can sequester cations such as Ba, Sr, and fission product oxides (~15 wt%), U, Cr, and Ni in the Ca sites (dark circles). Tetrahedra are Si and octahedral are Ti.	
Britholite (silicate apatite) Also known as oxy-apatites in the literature. [54, 82, 87, 315–318]	(REE, $Ca)_5(SiO_4,PO_4)_3(OH,F)$; i.e. $Ca_2Nd_8(SiO_4)_6O_2$, $Ca_2La_8(SiO_4)_6O_2$; based on ionic radii of Nd^{3+}, La^{3+}, and Pu^{3+}, an extensive range of solubility for Pu^{3+} substitution for the Nd or La, particularly on the *6h* site, is expected. Since there is an extensive range in the Ca/RE ratio in these silicate apatites, a fair amount of Pu^{4+} substitution may be possible; La^{3+} through Lu^{3+} can substitute for Ca^{2+} and form oxyapatites, $RE_{4.67}\square_{0.33}[SiO_4]_3O$; can also accommodate Sr and Cs, Th, U, Np.	 [from 319]

Framework silicates

Zeolites [320–325]	$(X_{x/n}[(AlO_2)_x\ (SiO_2)_y]$, where X is the charge balancing counter-ion, n is the charge of the counter-ion, x is the number of charge-deficient alumina sites, and y is the number of charge-neutral silica sites; characterized by internal voids, channels, pores, and/or cavities of well-defined size in the nanometer range, ≈ 4–13 Å; channels and/or cavities may be occupied by charge-compensating ions and water molecules. Zeolites like Ag-Mordenite selectively sorbs I_2 (^{129}I); certain zeolites can be converted to condensed oxide ceramics by heating. This process is particularly attractive for waste form fabrication because capture and storage is performed with minimal steps.
Pollucite [327–332]	$(Ca, Na)_2Al_2Si_4O_{12}•2H_2O$; host for fission products such as ^{137}Cs
Pollucite Cs/Ti Version [333–342]	$CsTiSi_2O_{6.5}$

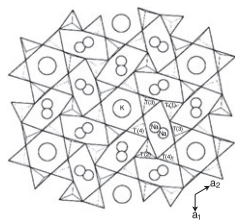
Structure of Zeolite-A [326] showing alternate Al and Si atom ordering but omitting the tetrahedral oxygens around each Al and Si.



[from 357]

Continued

Table 6.8 Continued

Crystalline ceramic phase	Comments	Structure
Nepheline [49, 343–348]	$NaAlSiO_4$ silica 'stuffed derivative' ring-type structure; some polymorphs have large nine-fold cation cage sites while others have 12-fold cage-like voids that can hold large cations (Cs, K, Ca). Natural nepheline structure accommodates Fe, Ti and Mg.	 Two-dimensional representation of the structure of nepheline showing the smaller eight oxygen sites that are occupied by Na and the larger nine oxygen sites that are occupied by K and larger ions such as Cs and Ca. [357]
Leucite*	$KAlSi_2O_6$; K analogue of nepheline	

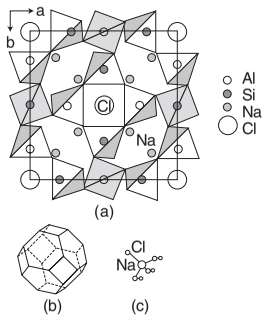
Sodalite group (name of mineral changes with anions sequestered in cage structure) [49, 343, 347–356]

Sodalite $Na_8Cl_2Al_6Si_6O_{24}$ also written as$(Na,K)_6[Al_6Si_6O_{24}]\bullet(2NaCl)$ to demonstrate that 2Cl and associated Na atoms are in a cage structure defined by the aluminosilicate tetrahedra of six adjoing $NaAlSiO_4$; a naturally occurring feldspathoid mineral; incorporate the alkali, alkaline earths, rare earth elements, halide fission products, and trace quantities of U and Pu (sodalite was and is being investigated as a durable host for the waste generated from electro-refining operations deployed for the reprocessing of metal fuel); minor phases in high level waste (HLW) supercalcine waste forms* where they retained Cs, Sr, and Mo, e.g. $Na_6[Al_6Si_6O_{24}](NaMoO_4)_2$; sodalite structures are known to retain B, Ge, I, Br, and Re in the cage-like structures

Nosean, $(Na,K)_6[Al_6Si_6O_{24}](Na_2SO_4)$), silica 'stuffed derivative' sodalite cage-type structure host mineral for sulfate or sulfide species.

Hauyne, $(Na)_6[Al_6Si_6O_{24}]((Ca,Na)SO_4)_{1-2}$ sodalite family; can accommodate either Na_2SO_4 or $CaSO_4$

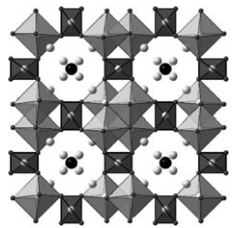
Helvite $(Mn_4[Be_3Si_3O_{12}]S$: Be can be substituted in place of Al and S_2 in the cage structure along with Fe, Mn, and Zn



Structure of Sodalite showing (a) two-dimensional projection of the (b) three-dimensional structure and (c) the fourfold ionic coordination of the Na site to the Cl^- ion and three framework oxygen bonds [357].

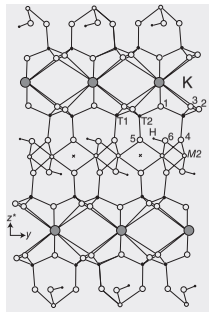
Continued

Table 6.8 Continued

Crystalline ceramic phase	Comments	Structure
	Danalite ($Fe_4[Be_3Si_3O_{12}]S$)	
	Genthelvite ($Zn_4[Be_3Si_3O_{12}]S$)	
	Lazurite, $(Ca,Na)_6[Al_6Si_6O_{24}]((Ca,Na)S,SO_4,Cl)_x$; can accommodate either SO_4 or S_2,Ca or Na and Cl	
Cancrinite [358]	$(Na,Ca,K)_6[Al_6Si_6O_{24}]((Na,Ca,K)_2CO_3)_{1.6}\bullet 2.1H_2O$ only found in hydroceramic waste forms	
Crystalline SilicoTitanate (CST) [336, 359–363]	$[(Ca,Na,K,Ba)AlSiO_4$ incorporates Ca, Na, K, Ba, Cs, and Sr	 Crystal structure of Cs exchanged Nb–titanium silicate. The dark and light grey spheres represent Cs^+ cations and water molecules, respectively [from 364].

Micas (dehydroxylated) [365–367]

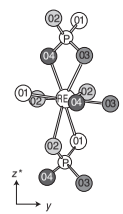
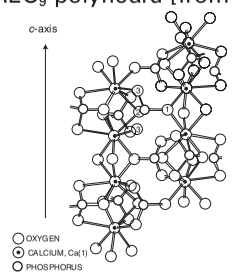
The following dehydroxylated micas have been synthesized phase pure: $LiAl_3Si_3O_{11}$, $NaAl_3Si_3O_{11}$, $KAl_3Si_3O_{11}$, $RbAl_3Si_3O_{11}$, $CsAl_3Si_3O_{11}$, $TlAl_3Si_3O_{11}$, $Ca_{0.5}\square_{0.5}Al_3Si_3O_{11}$, $Sr_{0.5}\square_{0.5}Al_3Si_3O_{11}$, $Ba_{0.5}\square_{0.5}Al_3Si_3O_{11}$, $La_{0.33}\square_{0.66}Al_3Si_3O_{11}$. In the Cs-mica up to 30 wt% Cs_2O can be accommodated, in the Rb-mica up to 22 wt% Rb_2O can be accommodated, and in the Ba-mica up to 19 wt% BaO can be accommodated. Mg, Fe^{2+}, Fe^{3+}, Mn, Li, Cr, Ti and V can substitute for VI-fold coordinated Al^{3+}.

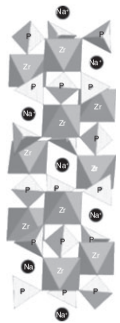
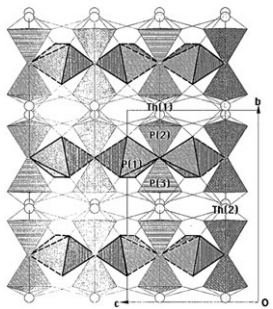


[from 368]

Continued

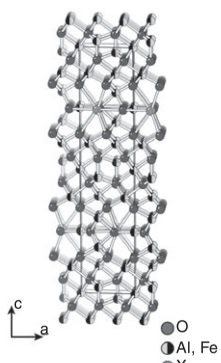
Table 6.8 Continued

Crystalline ceramic phase	Comments	Structure
Phosphates		
Monazite [218, 274, 369–373]	$CePO_4$ or $LaPO_4$; very corrosion resistant and can incorporate a large range of radionuclides including actinides and toxic metals into its structure; it has been proposed as a potential host phase for excess weapons plutonium and as a host phase for radionuclides and toxic metals in glass-ceramic waste forms for low-level and hazardous wastes.	 Alternating chains of PO_4 tetrahedra and REO_9 polyhedra [from 374].
Xenotime [218]	YPO_4	
Apatite [10, 54, 45, 218, 274, 317, 318, 372, 375–384]	$Ca_{4-x}RE_{6+x}(SiO_4)_{6-y}(PO_4)_y(O,F)_2$; actinide-host phases in HLW glass, glass-ceramic waste forms, ceramic waste forms and cement; actinides can readily substitute for the rare earth elements in the crystal structure, as in $Ca_2(Nd,Cm,Pu)_8(SiO_4)_6O_2$, and fission products are also readily incorporated. However, the solubility for tetravalent Pu may be limited without other charge compensating substitutions; has been proposed as a potential host phase for Pu and high-level actinide wastes.	 [from 385]

Sodium zirconium phosphate (NZP) [218, 386–391]	$NaZr_2(PO_4)_3$; structure can incorporate a complex variety of cations, including plutonium; three-dimensional network of corner-sharing ZrO_6 octahedra and PO_4 tetrahedra in which plutonium can substitute for Zr, as in $Na(Zr,Pu)_2(PO_4)_3$. Complete substitution of Pu^{4+} for Zr has been demonstrated in NZP. Cs and Sr can substitute for Na while fission products and actinides substitute for Zr in octahedral positions. P is tetrahedral.	 [from 388]
Thorium phosphate diphosphate (TPD) [218, 392–394]	$Th_4(PO_4)_4P_2O_7$; a unique compound for the immobilization of plutonium and uranium; partial substitution of Pu for Th has been demonstrated (up to 0.4 mole fraction), complete substitution is not possible.	 [from 395]

Continued

Table 6.8 Continued

Crystalline ceramic phase	Comments	Structure
Aluminates		
Magnetoplumbites [22, 62, 396–399]	Nominally $X(Al,Fe)_{12}O_{19}$, where X = Sr,Ba,$(Cs_{0.5} + La_{0.5})$ and $(Na_{0.5} + La_{0.5})$. The X site is XII-fold coordinated and both Cs^{+}/Ba^{2+}-Fe^{3+}/Fe^{2+} or Cs^{+}/Ba^{2+}-Ti^{4+}/Ti^{3+}-type substitutions can occur. Accommodating structures because they are composed of spinel blocks with both IV-fold and VI-fold coordinated sites for multivalent cations and interspinel layers which have unusual V-fold sites for small cations. The interspinel layers also accommodate large cations of 1.15–1.84 Å, replacing oxygen in XII-fold sites in the anion close packed structure. The large ions may be monovalent, divalent, or trivalent with balancing charge substitutions either in the interspinel layer $(Na_{0.5} + La_{0.5})$ or between the interspinel layer and the spinel blocks $(Cs^{+}/Ba^{2+}$-Fe^{3+}/Fe^{2+} or Cs^{+}/Ba^{2+}-$Ti^{4+}/Ti^{3+})$.	

* Supercalcines were the high temperature silicate-based 'natural mineral' assemblages proposed for HLW waste stabilization in the United States (1973–1985).

Adapted from [11].

Table 6.9 Attributes and examples of encapsulant and embedded waste forms (cements, geopolymers, ceramicrete, hydroceramics, and bitumen)

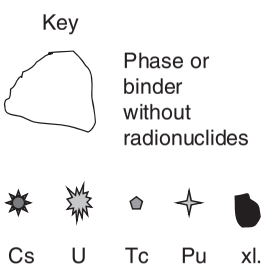
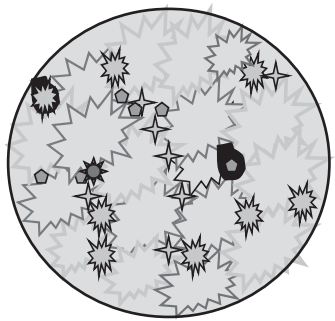
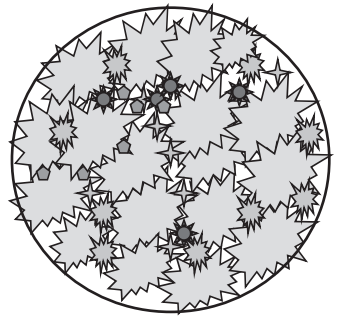
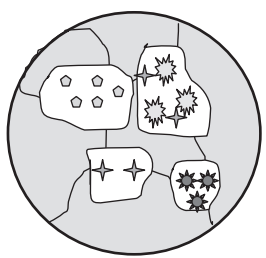
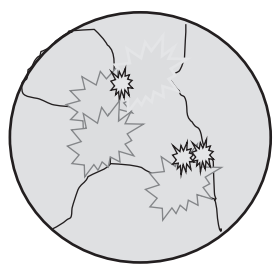
Waste form(s)	Encapsulated waste forms	Embedded waste forms
Description	Liquid waste is mixed with concrete or other binder – hydrated phases occur that can incorporate the radionuclides or hazardous species weakly or retain them by sorption. Example Cs and U sequestered by C-S-H hydrates and U and Tc sequestered by secondary fly ash granules. The remaining species are trapped on the grain boundaries of the interlocking C-S-H phases.	Liquid waste is mixed with concrete of other binder – primary phases and any secondary phases created by hydration (if an active mechanism) do not retain or sorb the radionuclide or hazardous species. Example shows Tc, Cs, U, and Pu all on the grain boundaries.
Radionuclide immobilization mechanism	Encapsulation and some chemical incorporation	Encapsulation and no chemical incorporation
		
		
Waste loading(s)/durability	(a) low waste loadings, (b) lower overall durability, (c) difficult to model radionuclide release from hydrated secondary phases and grain boundaries, (d) easy to process – usually mix and cast	(a) low waste loadings, (b) lower overall durability, (c) difficult to model radionuclide release from grain boundaries, (d) easy to process – usually mix and cast, (e) in case of bitumen, must be heated to flow so embedding can occur
Immobilization technologies	Mix and pour (cement, geopolymer, ceramicrete), Heat, mix, and pour (bitumen), Mix, pour, cure at slightly elevated temperatures (hydroceramics)	

Table 6.10 Attributes and examples of composite waste forms

Waste form(s)	Composite	Composite
Description	**Multiphase granular oxides/minerals/metals** (must be monolithed due to disposal requirements if not containerized)	**Previously made waste forms in need of remediation** (monolithing agents can be numerous and include glass – see GCMs above)
Radionuclide immobilization mechanism	Chemical incorporation and encapsulation/embedding	Chemical incorporation and encapsulation/embedding
Key: Phase or binder without radionuclides; Cs U Tc Pu xl.		
Waste loading(s)/ durability	(a) high waste loadings only if binder (monolithing agent) is minimized, (b) superior overall durability – double containment, (c) difficult to model radionuclide release from multiple phases, (d) need to tailor for and determine radionuclide partitioning amongst phases, (e) may require precalcining for certain processes to work efficiently	(a) high waste loadings only if binder (monolithing agent) is minimized, (b) superior overall durability – double containment, (c) difficult to model radionuclide release from multiple phases, (d) need to determine radionuclide partitioning amongst phases
Immobilization technologies for matrix phase	Mix and pour (cement, geopolymer, ceramicrete), Heat and pour (glass or metal), Heat, mix, and pour (bitumen), Mix, pour, cure at slightly elevated temperatures (hydroceramic)	

in simulated waste glasses (Table 6.4). These more highly ordered or polymerized regions of MRO, often have atomic arrangements that approach those of crystals and are often referred to as quasi-crystalline species or quasi-crystals. Crystalline ceramics incorporate radionuclides and hazardous species by a combination of SRO, MRO, and LRO. The LRO defines the periodic structural units characteristic of crystalline ceramic structures. In glass, glass-ceramics, glass composite materials (GCMs), and crystalline ceramics, the radioactive and hazardous constituents are atomically bonded by a combination of SRO, MRO, and LRO. In GCMs there is additional encapsulation of the ceramic components in the glass matrix.

6.4.1 Solidification by chemical incorporation

Vitrification

Vitrification is currently the most widely used technology for the treatment of high level radioactive wastes (HLW) throughout the world (Tables 6.1, 6.3 and 6.4). Development of glasses for the solidification of HLW began at different times in the US, Canada, Europe, and the USSR [17]. Different glass formulations (borosilicate, aluminosilicate, and phosphate glasses) and processing strategies were developed [18]. Currently, most of the nations that have generated HLW are immobilizing in either borosilicate glass or aluminophosphate glass. One of the primary reasons that glass has become the most widely used immobilization media is the relative simplicity of the vitrification process, e.g. melt waste plus glass forming additives and cast. There is >50 years processing experience[5] with commercial borosilicate glasses and borosilicate glasses have favorable systems evaluations in terms of both melting and product behavior.

Melting homogenizes the mixture and so this process is easier to perform remotely than a ceramic waste form process that requires powder handling, e.g. mechanical mixing of waste and ceramic additives and grinding for particle size control, followed by cold pressing and sintering or hot pressing at elevated temperatures. A second reason that glass has become widely used for HLW is that the amorphous and less rigid structure of glasses (SRO and MRO) compared to ceramics (SRO, MRO, and LRO) enables the incorporation of a very large range of elements that are atomically bonded in the flexible glass structure (see Table 6.4). Thus glasses can accommodate larger waste composition fluctuations than most ceramics.

The glass forming SRO structural groups are usually tetrahedral Si, B, Al, Fe, P surrounded by four oxygen atoms (tetrahedral coordination) or B

[5]Phosphate glasses (aluminophosphates and iron phosphates) are not used commercially as frequently as the borosilicates and hence are not as well studied in HLW stabilization applications.

surrounded by three oxygen atoms (trigonal coordination) and glasses are named after the predominant tetrahedral species, e.g. borosilicates have primarily B and Si with some Al, Fe, and P and aluminophosphates would have primarily Al, P, and Si. See Table 6.4 for the attributes of various types of glasses that have been used for a variety of HLW wastes and pertinent references that can be consulted.

The tetrahedra and trigonal species in glass link to each other via bridging oxygen bonds (BO). The remaining non-bridging (NBO) atoms carry a negative charge and, in turn, ionically bond to positively charged cations like Cs^{+}, Sr^{+2}, Ca^{+2} and positively charged waste species. These linkages create the MRO structural groups such as (Cs,K,Na,Li)AlO_2, (Cs,K,Na,Li)FeO_2, (Cs,K,Na,Li)BO_2, and (Cs,K,Na,Li)SiO_4 [19] or (Cs,K,Na)$AlSiO_4$ [20] which form sheet-like units, chain-like units, and monomers [21] that further bond the waste species ionically.

The tetrahedra define the network regions, while NBO define percolation channels or depolymerized regions (DR) shown in Table 6.4 that can act as ion-exchange paths for elements that are less well bonded to the NBO. Such percolation channels are also found in rare-earth (lanthanide) alumino-borosilicate (LaBS) glasses as well (see Table 6.4). Thus, the molecular structure of glass controls radionuclide/contaminant release by establishing the distribution of ion exchange sites, hydrolysis sites, and the access of water to those sites through the percolation channels, and the mechanisms are similar to those observed in natural analog glasses (basalts) and in mineral analogs.

Moreover, HLW glasses melt at lower temperatures (1050–1150°C) than higher ceramic waste forms, which minimizes the volatility of radioactive components such as ^{99}Tc, ^{137}Cs, and ^{129}I. While ceramics are often credited with having higher chemical durability than glasses, if the radionuclides are incorporated in an intergranular glassy phase during processing (see discussion in the next section), they leach at about the same rates as those from glassy wasteforms [22].

Lastly, nuclear waste glasses have good long-term stability including irradiation resistance and excellent chemical durability. In addition, the ease of modeling the durability of a homogeneous rather than a heterogeneous material in terms of having only one radionuclide source term is also an advantage.

A basic assumption in all glass dissolution models is that the solid being modeled is made up of a single phase and so the durability response has only one source term (see Table 6.3). Therefore, phase separated glasses (with two source terms) with two distinct glass compositions are avoided as their durability cannot currently be modeled. Often the two immiscible glass phases have different compositions, e.g. one phase is often boron-rich and has a poorer durability than the bulk and/or the matrix phase. Having

a poorly soluble second phase is not desirable for HLW glasses where the distribution of the radionuclides in the two glassy phases would have to be known for every waste glass fabricated. Since the volume fraction of each phase is also related to the thermal history of each canister of glass, each canister would be different and this complicates durability modeling to the point that it is virtually impossible.

To ensure that HLW borosilicate glasses are homogeneous (not phase separated), a minimum Al_2O_3 limit is applied in the US. The effect of insufficient Al_2O_3 was first reported by French researchers [23] who determined that many glass durability models were non-linear, e.g., glasses had release rates far in excess of those predicted by most models, in regions corresponding to low Al_2O_3 and in excess of 15 wt% B_2O_3. The phenomenon was independently discovered by US researchers and found to exist in natural basalt glass systems as well [24–26].

Additional durability source terms can occur if crystals are present in a glass because crystals create grain boundaries that can (1) selectively undergo accelerated dissolution while the crystals themselves may have a different dissolution response [27], or (2) have compositions not representative of the bulk glass [28]. This will be discussed further in the next section on glass ceramics.

Glass formulations are generally homogeneous, allowing only a few weight percent crystals to form on cooling in the canister. Certain crystals such as iron spinels have little impact on glass durability as they are themselves very durable and cause minimal grain boundary dissolution [27, 29]. However, for other phases such as nepheline, acmite, and lithium silicates that are less durable than iron spinels and not isotropic, the impact on glass durability from the crystal and the grain boundaries can be pronounced. This is especially true if the crystal sequesters radionuclides as this gives a secondary source term for radionuclide release. Therefore, durability testing must be performed to confirm that any crystallization that might occur during canister cooling has minimal impact [30–33]. This ensures that the last three terms in Eq. [6.1] approximate zero and that glass dissolution has a single source for radionuclide and hazardous species release

$$\sum \text{Durability} = \underbrace{\text{durability}_{(\text{homogeneous})}}_{\text{1st term}} + \underbrace{\text{durability}_{(\text{amorphous phase separation})}}_{\text{2nd term}} + \underbrace{\text{durability}_{(\text{crystallization})}}_{\text{3rd term}} + \underbrace{\text{durability}_{(\text{accelerated grain boundary})}}_{\text{4th term}} \quad [6.1]$$

This durability equation will be discussed in more detail in reference to other waste forms where the third and fourth terms in Eq. [6.1] may become important.

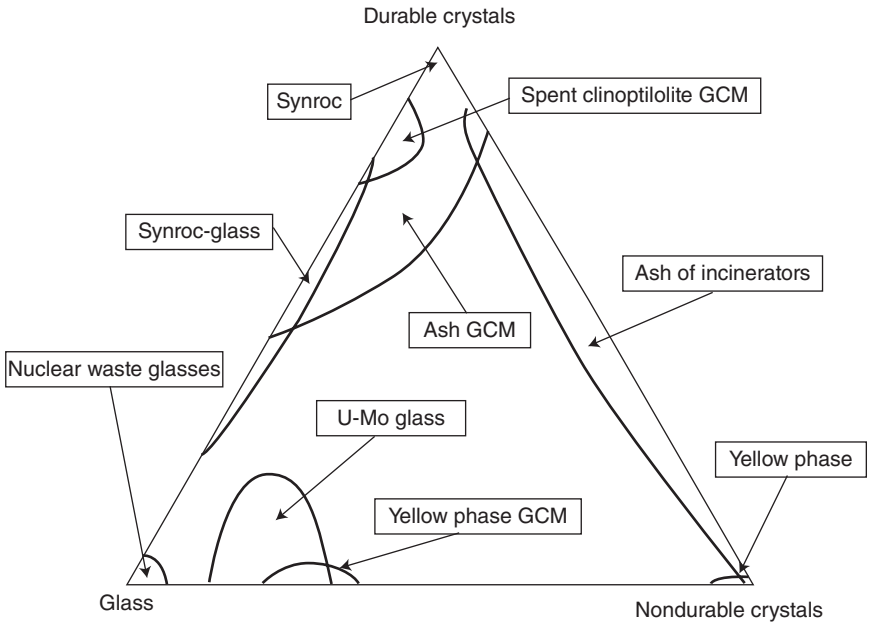
Glasses can be made by JHM, AJHM, induction melting, CCIM, and HIPing (see Table 6.3). Extensive reviews on vitrified waste forms can be found in the references cited in Section 6.8 and in Table 6.4.

Glass ceramics and glass composite materials (GCMs)

It is sometimes difficult to distinguish between glass-ceramics (a glassy matrix which is allowed to form crystals during cooling or glassy matrices where controlled cooling is used so that certain crystalline species known to sequester radionuclides are encouraged to form) and glass composite materials (GCMs) (Tables 6.5 and 6.6) [34, 35]. GCMs are considered a composite material where the long-lived radionuclides are atomically bonded in the ceramic (mineral) phase and the glass is an encapsulating matrix phase. The glass can have little or no retention of radionuclides or act as the host for the short-lived radionuclides [36]. Glass-ceramics and GCMs include glass-ceramics where a glassy waste form is crystallized in a separate heat treatment, GCMs formed by melt crystallization (controlled or uncontrolled), and GCMs in which a refractory waste is encapsulated in glass [34]. Glass-ceramics and GCMs offer increased waste loadings, increased waste form density, and thus smaller disposal volumes.

One such example of a GCM in Table 6.6 is the glass bonded sodalite, as the radionuclide of concern, ^{129}I, is in the ceramic phases and not in the glassy phase. Other examples of GCMs include the following [36]:

1. glass ceramics in which a glassy waste form is crystallized in a separate heat treatment [7, 37];
2. GCMs in which, for example, a refractory waste is encapsulated in glass such as hot-pressed lead silicate glass matrix encapsulating up to 30 vol% $La_2Zr_2O_7$ pyrochlore crystals to immobilize minor actinides [38];
3. GCM formed by pressure-less sintering of spent clinoptiloite from aqueous waste processing [39];
4. some difficult wastes such as the French HLW U/Mo-containing materials immobilized in a GCM termed U-Mo glass formed by cold crucible melting that partly crystallize on cooling [40];
5. 'yellow phase' containing wastes are immobilized in Russia in a 'yellow phase GCM' containing up to 15 vol% of sulfates, chlorides, and molybdates [41]; and
6. GCM that immobilizes ashes from incineration of solid radioactive wastes [42].



6.1 Current homogeneous glass formulations are limited to the lower left-hand corner of this triangular diagram. If the homogeneous glasses crystallize durable crystals shown at the apex of the triangle, e.g. spinels, ZrO_2, apatite, TiO_2, etc., then waste loading can be increased and glass composite materials (GCMs) produced by changing the melter technology (e.g. CCIMs) or invoking a different technology such as HIPing. Ceramic waste forms are at the apex and are considered exceptionally durable waste forms but may be more appropriate as small volume waste forms as processing is more difficult. Some ceramic waste form formulations can be melted in advanced melters like CCIMs and then allowed to crystallize into GCMs. While certain species such as Mo, S, and P can create non-durable secondary phases (lower right apex of the triangle), these should be avoided or macroencapsulated, which moves their durability closer to the lower left apex of the triangle [36].

Note that alkali-rich wastes at the Hanford site that were made by in-container vitrification (ICV)[6] produced an immobilized glassy waste form with high crystal content that characterize them as GCMs [43].

Note that yellow phase is composed of species that are poorly soluble in glass such as Na_2SO_4 which can sequester Cs and Sr [44], (Na,K,Cs)Cl, $(Na,K,Cs)_2Cr_2O_7$, and $(Na,K,Cs)_2MoO_4$. Yellow phase is either (1) prevented from crystallizing (Fig. 6.1) or (2) the glass is heat treated to

[6]This is not the baseline AJHM process that will produce a homogeneous glass with minimal crystallization.

encapsulate the soluble phase(s) as GCMs. One such vitrification process given as the fifth example above produces a sulfate-chloride-molybdate GCM by using vigorous melt agitation followed by rapid cooling of the melt to the upper annealing temperature to fix the dispersed 'yellow phase' into the host borosilicate or aluminosilicate glass. The sulfate–chloride-molybdate-containing GCM (see yellow phase GCM in Fig. 6.1) has only a slightly diminished chemical durability compared with sulfate–chloride-molybdate-free aluminosilicate and borosilicate glasses [36].

In many cases, until a waste form is made and analyzed for the distribution of radionuclides amongst the crystalline and glassy phases, one cannot discern whether a GCM has been made (see Table 6.5). In either case, glass-ceramics and GCMs offer a useful compromise between glasses and ceramics, being easier and less expensive to prepare than conventional ceramics, but offering higher durability than glasses.

Depending on the intended application, the major component may be a crystalline phase with a vitreous phase acting as a bonding agent, or, alternatively, the vitreous phase may be the major component, with particles of a crystalline phase dispersed in the glass matrix. Glass-ceramics and GCMs may be used to immobilize long-lived radionuclides (such as actinide species) by incorporating them into the more durable crystalline phases, whereas the short-lived radionuclides may be accommodated in the less durable vitreous phase [36].

Historically, crystallization of vitreous waste forms has been regarded as undesirable as the crystallization has the potential to alter the glass composition and hence the durability of the remaining continuous glass phase could eventually be compromised when it comes into contact with water. However, there has been a recent trend towards higher crystallinity in vitreous waste forms so that they are more correctly termed glass-ceramics or GCMs depending on whether the glass or the crystals contain the radionuclides (Table 6.5).

Table 6.5 also shows glass-ceramics where significant quantities of crystals (arising from higher waste loadings) form, such as in the Savannah River Site (SRS) high iron bearing glasses where spinel crystallizes [27] and the crystals do not incorporate the radioactive species but act as benign or inert 'stones' in the glass.

Historically silicate glass-ceramics were developed in the mid 1970s in Germany [45]. Silicate and phosphate glass-ceramics were also developed in the USSR [46], silicate glass-ceramics were developed in Japan [47], and titanium aluminosilicate glass-ceramics were developed in Canada [48]. GCMs represent a second generation, more sophisticated approach to the production of glass-ceramics, where the long-lived radionuclides are forced into the more durable crystalline phases by tailoring the waste-additive mixture and/or controlling the crystallization. More recently, GCMs such

as the glass bonded ceramic waste forms containing sodalite and alkali halides in a borosilicate matrix have been developed for electrorefiner wastes, specifically the stabilization of ^{129}I in sodalite and NaI. [49–51], while rare-earth oxyapatites, powellite, celsian, and pollucite [52] have been developed for rare-earth lanthanide and Cs, high Mo-containing wastes. Excellent reviews of other GCM's, such as SYROC glass ceramics, muratitie, and other Ti-based glass ceramics can be found in Stefanovsky *et al*. [3], Donald *et al*. [4, 8] and Lee *et al*. [34].

In terms of modeling the durability of glass-ceramics and GCMs, the distribution of the radionuclides amongst the crystalline and glassy phase becomes important. Referring back to Eq. [6.1] which gives the needed durability vectors for each phase, we see that the second term drops out since the glassy phase should not have glass-in-glass phase separation, leaving terms 1, 3, and 4 (Eq. [6.2]):

$$\sum \text{Durability} = \underbrace{\text{durability}_{(\text{homogeneous})}}_{\text{1st term}} + \underbrace{\text{durability}_{(\text{crystallization})}}_{\text{3rd term}} + \underbrace{\text{durability}_{(\text{accelerated grain boundary})}}_{\text{4th term}} \quad [6.2]$$

If the glass contains no radionuclides then the first term in Eq. [6.2] also drops out. If there are multiple types of phases present and each hosts a different radionuclide, then there will be durability vectors for each phase that hosts that radionuclide as shown in the Table 6.6 example for the ^{129}I in glass-bonded sodalite waste forms. If there are no radionuclides in the crystals then the third term drops out and it may be possible to demonstrate that the fourth term is negligible. If a given radionuclide is present in both the glassy phase and a crystalline phase, then the durability response from the glass and the crystalline phase and the grain boundary are additive as shown in Eq. [6.2].

Ceramic and mineral waste forms

The concept of immobilizing the radioactive elements of nuclear waste in an assemblage of mineral phases was originally introduced by Hatch [1] at Brookhaven National Laboratory in 1953 (Tables 6.7 and 6.8). The feasibility of making a ceramic of natural mineralogically stable phases was demonstrated by McCarthy [53, 54] and Roy [2, 55] at the Pennsylvania State University between 1973 and 1976. Since that time, a number of other mineralogic-ceramic assemblages have been developed. Among these high temperature (1000–1500°C) processes are the Sandia titanate-based ceramic [56], the Australian titanate-based ceramic 'Synroc' [57–59], the silicate-phosphate supercalcine ceramics [60], the alumina-based tailored ceramics

[61, 62], and the Pu pyrochlores [63]. Often in ceramics made by cold pressing and sintering or hot isostatic pressing, an intergranular glassy phase is produced during liquid phase sintering on the ceramic grain boundaries and the radionuclides preferentially migrate to the glassy phase(s) [64–72]. If the radionuclides are incorporated in the intergranular glassy phase(s), they have been found to leach at about the same rates as those from glassy waste forms [22].

Crystalline (ceramic/mineral) waste forms made by moderate temperature (700–750°C) thermal treatment have not been as intensely investigated as those formed at high temperatures as discussed above [61]. However, crystalline wasteforms made from clay have also been studied almost continuously since the work of Hatch in 1953 [1, 61]. Often the high temperatures used for sintering of supercalcine ceramics created sodalite-cancrinite mineral assemblages. In 1981, Roy [73] proposed low temperature hydrothermally processed low solubility phase assemblages consisting of the micas, apatite, pollucite, sodalite-cancrinite, and nepheline, many of which could be made from reaction of various clays (kaolin, bentonite, illite) with waste.

Clay-based crystalline (ceramic/mineral) waste forms were not pursued in the late 1970s and early 1980s because there was no continuous commercial technology available that could process the waste/clay mixtures in a hydrothermal environment [61]. A commercial facility to continuously process radioactive wastes by pyrolysis at moderate temperatures in a hydrothermal steam environment was built by Studsvik in Erwin, Tennessee in 1999 [74, 75]. This facility uses a fluidized bed steam reforming (FBSR) technology to pyrolyze ^{137}Cs and ^{60}Co organic resins from commercial nuclear facilities. This technology has the capability to process a wide variety of solid and liquid streams including wastes containing organic ion exchange resins, charcoal, graphite, sludge, oils, solvents, and cleaning solutions at radiation levels of up to 400 R/hr. When clay is added as a mineralizing agent, the feldspathoid minerals (sodalite, nosean and nepheline) are formed by nanoscale reaction with the clay. The phases formed act as hosts for high Cl, I, F, ^{99}Tc, and SO_4 alkali (Na, K, Cs) bearing wastes [76–80] and organics are destroyed creating steam and CO_2. The mineralization occurs at the moderate FBSR temperatures because the FBSR operating temperature is in the range in which most clays become amorphous at the nanoscale level, e.g. kaolin, bentonite (montmorillonite), and illite. The clays lose their hydroxyl (OH^-) groups at the FBSR temperatures which destabilizes the octahedral (six nearest neighboring atoms that form an octagon) Al^{3+} cation in their structure (Fig. 6.2) and they become amorphous as confirmed by X-ray diffraction (XRD) analysis. The alkali in the waste 'alkali activates' the unstable Al^{3+} cation to form new mineral phases and the fluidizing agent, steam, catalyzes the mineralization. In the absence of steam, many of these mineral phases only form at temperatures of >1200°C.

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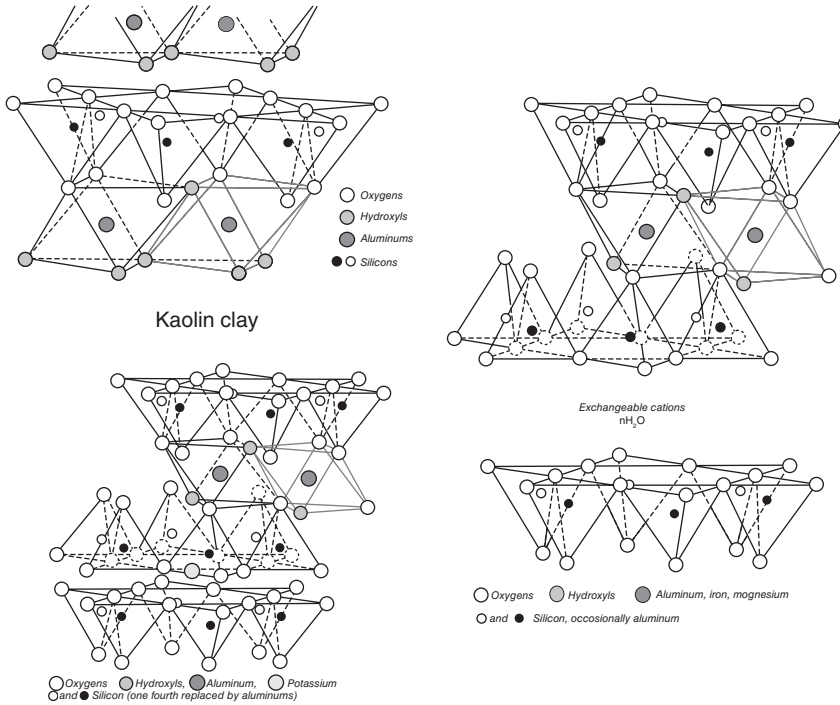


6.2 Atomic structures of various clays (kaolin, bentonite-montmorillonite, illite). After [400, 401].

Ceramic waste forms can be single phase, e.g. UO_2, or single-phase solid solutions like $(U,Th,Pu)O_2$ (Table 6.7). Multiphase ceramics are formulated so that each radionuclide can substitute on a given host lattice in the various phases (see Table 6.8).

Of great importance when relying on the LRO (size and coordination of the crystallographic site which will act as host to a given radionuclide or its decay product upon transmutation) is that the crystal-chemical substitutions must be electrically balanced [81, 82]. When a monovalent cation transmutes to a divalent cation, the substitutions must be coupled to retain the electrical balance of the host phase without destroying the integrity of the phase: the lattice site must be of suitable size and bond coordination to accept the transmutation. The bonding in crystalline ceramic or mineral phases can only maintain charge balance in one of two ways: (1) if sufficient lattice vacancies exist or (2) if a variable valance cation like Fe or Ti is present in a neighboring lattice site for charge balance. Both scenarios assume that the variable valence cations do not change lattice sites and that

the charge balancing cations are in the same host phase in nearby lattice sites. The lattice site must be of sufficient size or flexible enough to accommodate the transmuting cation. It is advantageous if the lattice site of the desired host phase has irregular coordination or is distorted as will be shown in some examples below.

The solubility or flexibility of a ceramic or mineral phase(s) as hosts for a substituted cation of a different valence can be studied by performing coupled substitutions on the phase pure mineral host phase. If the number of cations changes during the substitution, a vacancy is either created or consumed and the substitution must maintain electrical neutrality. These types of substitution are most often seen in polymorphic substitutions [83] of the type

$$\square + Ba^{2+} \rightarrow 2K^{+}$$

$$\text{or } \square + Ca^{2+} \rightarrow 2Na^{+}$$

$$\text{or } \square + Na^{+} + 2Ca^{2+} \rightarrow 3Na^{+} + Ca^{2+}$$

where □ denotes a vacancy. Implicit in these coupled substitutions is the fact that the exchanging cations occupy the same lattice sites, have the same coordination, and thus the crystallographic symmetry is maintained. Therefore, substitutions as described above should be written with roman numerals that designate the number of oxygen atoms that coordinate around a given cation, e.g. ^{VIII}Ca designates the octahedral VIII-fold coordination for the Ca^{2+} lattice site in oxyapatites:

$$\underbrace{3^{VIII}Ca^{2+}}_{\text{host phase}} \rightarrow \underbrace{2^{VIII}Nd^{3+}}_{\text{substituted phase}}$$

Calcium-neodymium coupled substitutions have been successful [81, 82] in the oxyapatite ($Ca_6[SiO_4]_3$) structure forming completely substituted $Nd_4\square_2[SiO_4]_3$ where two-thirds of the lattice sites have Nd^{3+} and one-third are vacant. In the oxyapatite structure, the Ca^{2+} is normally in VIII-fold coordination and has a 1.12 Å [84–86] atomic radius. The Nd^{3+} cation in VIII-fold coordination also has an atomic radius of 1.11 Å [86] very close to the Ca^{2+} atomic radius in VIII-fold coordination. Felsche showed that the rare earth elements La^{3+} through Lu^{3+} can substitute for Ca^{2+} and form oxyapatites, $RE_{4.67}\square_{0.33}[SiO_4]_3O$ [87]; see Table 6.8]. McCarthy and Davidson [54] showed that even more complex, but coupled, substitutions were possible in the oxyapatite structure such as

$$\underbrace{6^{VIII}Ca^{2+}}_{\text{host phase}} \rightarrow \underbrace{1.7^{VIII}Nd^{3+} + 1.7^{VIII}Cs^{+} + 0.86^{VIII}Ce^{4+} + {}^{VIII}0.86Sr^{2+} + 0.88\square}_{\text{substituted phase}}$$

where the atomic radius, *r*, of Cs^{+} in VIII-fold coordination is 1.74 Å, Ce^{4+} in VIII-fold coordination is 0.97 Å, and Sr^{2+} in VIII-fold coordination

is 1.26 Å. In this case, small radii cations such as Ce^{4+} are mixed with large radii cations like Cs^{+} so that individual lattice sites can distort without perturbing the entire crystal structure. Note that the exchanging cations are always in the same lattice site of the same host phase [54, 81, 82, 87].

The substitutions such as given above for the oxyapatites were also demonstrated [81, 82] to be possible in many other Ca-bearing cementitious mineral phases such as larnite (Ca_2SiO_4 or β-C_2S), alite (calcium trisilicate or Ca_3SiO_5 or C_3S), C_3A ($Ca_3Al_2O_6$), and C_4AF ($Ca_4Al_2Fe_2O_{10}$). This allowed Jantzen *et al.* [88, 89] to make substitutions for Ca^{2+} in each phase (up to ~15 mole%) and the following additional substitutions[7]:

$$\underbrace{Ca^{2+} + \square}_{\text{host phase}} \rightarrow \underbrace{2Cs^{+}}_{\text{substituted phase}}$$

$$\underbrace{2Ca^{2+}\square}_{\text{host phase}} \rightarrow \underbrace{Cs^{+} + {Sr_{0.5}}^{2+} + {Nd^{3+}}_{0.17} + {Ce^{4+}}_{0.25} + 0.08\square}_{\text{substituted phase}}$$

$$\underbrace{1.5Ca^{2+} + Si^{4+}}_{\text{host phase}} \rightarrow \underbrace{Sr^{2+} + Mo^{5+} + 0.5\square}_{\text{substituted phase}}$$

$$\underbrace{4Ca^{2+} + Fe^{3+} + Al^{3+}}_{\text{host phase}} \rightarrow \underbrace{0.66Nd^{3+} + Zr^{4+} + Mo^{4+} + Sr^{2+} + Ba^{2+} + 1.33\square}_{\text{substituted phase}}$$

$$\underbrace{\underbrace{4^{IX}Ca^{+2}}_{r\sim 1.18\ \text{Å}} + \underbrace{2^{VI}Fe^{+3}}_{r=0.65\ \text{Å}}}_{\text{host phase}} \rightarrow \underbrace{\underbrace{2.66^{IX}Nd^{+3}}_{r=1.16\ \text{Å}} + \underbrace{0.38^{VI}Ce^{+4}}_{r=0.87\ \text{Å}} + \underbrace{0.56^{VI}Zr^{+4}}_{r=0.72\ \text{Å}} + \underbrace{0.75^{VI}Fe^{+3}}_{r=0.65\ \text{Å}} + 1.65\square}_{\text{substituted phase}}$$

These types of crystal-chemical substitutions have been studied in (1) Synroc (Synthetic rock) titanate phases such as zirconolite ($CaZrTi_2O_7$), perovskite ($CaTiO_3$), and hollandites (nominally $Ba(Al,Ti)_2Ti_6O_{16}$) [90], and (2) in high alumina tailored ceramic phases such as magnetoplumbites (Table 6.8). The magnetoplumbites (discussed below) are also found as a minor component in Synroc when the waste being stabilized is high in Al [91].

In the Synroc phase assemblages, the hollandite phase is the Cs^{+} host phase. The structure can be written as $Ba_xCs_y(Al,Fe)_{2x+y}Ti_{8-2x-y}O_{16}$ where x + y must be <2 [92]. There are two types of octahedral sites. One accommodates trivalent cations like Al^{3+}, Ti^{3+}, and Fe^{3+}, while the other accommodates Ti^{4+}. The Cs^{+} is accommodated in tunnels that normally accommodate the Ba^{2+} cation. The Cs-Ba lattice sites are VIII-fold coordinated [90, 92].

[7]Note that the number of lattice sites have to be equivalent on the left-hand side and right-hand sides of the equation.

The substitution is ordered upon fabrication and incommensurate superstructures result when Cs^{+} substitutes for Ba^{2+} [91].

Cesium has been experimentally substituted for Ba when Fe^{3+} is substituted for Ti^{3+} in the VI-fold sites of hollandite. The species $\underbrace{^{VIII}Cs^{+}_{0.28}\,^{VIII}Ba^{2+}_{1.00}}_{\text{A site}}\,\underbrace{^{VI}Al^{3+}_{1.46}\,^{VI}Fe^{3+}_{0.82}}_{\text{B site}}\,\underbrace{^{VI}Ti_{5.72}O_{16}}_{\text{C site}}$ has been fabricated by sintering in air at 1320°C [92]. A Ba-Al hollandite ($Ba_{1.16}Al_{2.32}Ti_{5.68}O_{16}$) was electron irradiated (1–2.5 MeV) and β-irradiated (4×10^{8} to 7×10^{9} Gy) and found to contain Ti^{3+} centres and O_2^{-} superoxide ions which confirmed the mechanism of charge balance during transmutation [92]. Theoretically, the limiting y value in hollandite is 0.81 Cs which corresponds to a 9.54 wt% waste loading of Cs_2O [93].

Single-phase and multiphase ceramics can be made by many of the thermal treatment technologies given in Table 6.2. Examples include melting in smelters instead of melters, cold pressing (CIP or CUP) and sintering, hot isostatic pressing (HIP), or hot uniaxial pressing (HUP). Mineral waste forms made from clays can be made by FBSR. The clay minerals act as a template: kaolin templates the feldspathoid minerals (sodalite and nepheline), while illite clays template the micas (see Table 6.8).

In terms of modeling the durability of multiphase ceramics, the distribution of the radionuclides amongst the crystalline phases and in any intergranular glassy phase is important. Referring back to Eq. [6.1], which gives the needed durability vectors for each phase, we see that the second term drops out since the glassy phase should not have glass-in-glass phase separation leaving terms 1, 3, and 4 (Eq. [6.3]) where the first term should have a minimal durability impact unless large concentrations of the intergranular glass exist or large amounts of radionuclides have been sequestered in the glassy phase compared to the ceramic phase.

$$\sum \text{Durability} = \underbrace{\text{durability}_{(\text{homogeneous})}}_{\text{1st term}} + \underbrace{\text{durability}_{(\text{crystallization})}}_{\text{3rd term}} + \underbrace{\text{durability}_{(\text{accelerated grain boundary})}}_{\text{4th term}} \quad [6.3]$$

If there are multiple types of phases present in the ceramic and each hosts a different radionuclide, then there will be durability vectors for the each phase that hosts that radionuclide as shown in the Table 6.6 example for the ^{129}I in glass-bonded sodalite waste forms.

Metals

A metal waste form (MWF) has been under development for stabilization of the metallic fuel hulls from spent nuclear fuel processed pyrochemically.

As the spent fuel is chopped, the fuel materials are removed by the pyrochemical processing and a stainless steel shell (called a hull or cladding hull) is left in the basket of the bath system. The process removes uranium, actinides, and most fission products, leaving behind the hulls, fuel alloy material (generally zirconium), and any noble metal fission products (like technetium) in the basket [94]. The noble metal fission products remain somewhat adhered to the surface of the stainless steel hulls and the hulls are coated with salt from the salt bath.

The basket is processed to remove the salt and solidify the hulls, alloy, and other metals into a consolidated waste form. The hulls are solidified by melting the metal into a uniform, homogeneous wasteform (1,560°C). Once homogeneous, the metal alloy should cool to a single phase. Typically, some zirconium (in addition to that remaining from the alloy) is added to bring the metal to about 15 wt% zirconium and lower the melting point of the mass. With the exception of the zirconium to control melting temperature, very few additives are made to the primary waste (cladding hulls), and the overall waste loading is typically above 90% [95].

The metallic waste seems to be a simple waste form with little development necessary. It has high waste loading, is durable, and fairly straightforward to process. The only development that might make a difference would be an evaluation of whether the cladding could be removed from the process before electrorefining and disposed of separately as a low-level waste form that is potentially greater than Class C. However, the cladding is the host form for the noble metal fission products (notably technetium), and separate disposition would probably require developing a different waste form for those radionuclides.

Likewise, MWF are under study by ANSTO for applications in the United Kingdom by HIPing. In this case, metal encapsulation is to be used for immobilizing debris waste streams that are uneconomical to handle separately, e.g. cermets, SiC, graphite, broken fuel pins, fuel hulls, etc. The process is the same as that used to make glass-ceramic and full-ceramic waste forms and so the processing method is multipurpose.

In spent nuclear fuel (SNF), epsilon metal (ε-metal) composed of Mo-Tc-Ru-Pd-Rh is generated from the fission process and heat. The ε-metal phase in SNF forms in the same manner that ε-metal formed in the natural reactors in Gabon, Africa some 2 billion years ago and has survived largely unchanged except for the decay of ^{99}Tc. Therefore, ε-metal has shown long-term stability in nature. This metal does not dissolve during the acid dissolution of SNF but forms solid particles with dimensions of ~10 μm in the dissolver sludge. This sludge was formed into a monolithic waste form, by arc melting at 1,800°C into an alloy pellet containing Ru, Re (substitute for ^{99}Tc), Mo, Pd, and Rh in the appropriate masses of each metal [96]. Dissolution rates of 4×10^{-5} g/(m^2 d) and 4×10^{-3} g/(m^2 d) were reported for synthetic

ε-metal phase and ε-phase harvested from SNF under reducing and oxidizing conditions in static durability testing [97, 98].

6.4.2 Solidification by encapsulation

This section primarily discusses non-thermal methods of encapsulation. The thermal encapsulation by glass is covered in Section 6.4.1 on glass ceramic materials on pages 214–217.

Cements including grouts

Stabilization and solidification with cement-based binders has been used to immobilize radioactive wastes since the beginning of the nuclear age. The process has been used to encapsulate solid waste, solidify liquid waste (including tritiated water), stabilize contaminated soils, stabilize tank-heel residues after tanks are emptied, and as low permeability barriers. Cements have also been used as binders and to encapsulate granular or cracked waste forms.

Cements microencapsulate wastes, although there is recent evidence that during hydration three binding mechanisms can also occur between the cement and metal ions in the waste [99–101]:

- precipitation of metal ions into the alkaline matrix as an oxide, mixed oxide, or as another discrete solid phase;
- adsorption or (co-)precipitation of metal ions onto the surface of cement minerals;
- incorporation of metal ions into hydrated cement minerals as they crystallize.

These mechanisms are shown as examples in Table 6.9; with the binding mechanisms (reaction of the waste with the cement or grout particles) shown as encapsulation and without the binding mechanism shown as embedding.

These processes are not mutually exclusive (so both encapsulation and embedding take place) and the above classification partly reflects slow kinetics; previously adsorbed species may be incorporated as mature cement pastes. Nevertheless, it does allow some generalized guidelines to be formulated. The solubility of discrete heavy metal solid phases is a limiting factor with regard to the second and third mechanisms [102], so that only ions that do not precipitate as basic oxides tend to be incorporated in, or surface adsorbed to, hydrated cement minerals to a significant degree.

The principal minerals available in the hydrated Portland cement matrix are calcium silicate hydrate (C-S-H, 50 wt%), portlandite ($Ca(OH)_2$, 20 wt%), and Ca aluminates. The most important Ca aluminates

are ettringite ($3CaO.Al_2O_3.3CaSO_4.32H_2O$, 4 wt%), calcium aluminate monosulphate ($3CaO.Al_2O_3.CaSO_4.12H_2O$, 7 wt%) and Ca carboaluminate ($3CaO.Al_2O_3.CaCO_3.11H_2O$, 7 wt%) [103]. Together they make up almost 90 wt% of the mineral suite in hydrated ordinary Portland cement (OPC) paste and thus, have the greatest potential for metal(loid)-ion binding. The relative importance of the above processes for selected metals can be found in a recent review [104].

OPC is the most common type of cement used for immobilizing liquid and wet solid wastes worldwide [6]. Composite cement systems were developed in the UK for ILW encapsulation using additional powders as well as OPC such as blast furnace slag (BFS) and pulverized fuel ash (PFA). These offered cost reduction, energy saving and potentially superior long-term performance. BNFL, for example, use a 9:1 ratio of BFS to OPC to reduce the heat of hydration, which for OPC cements, would otherwise limit container volumes. Large containers (see Fig. 6.3) can therefore be used safely without concern over heat from setting reactions causing water to boil off.

Modeling has shown that cements can be 'designed' to retain radioactive and hazardous constituents [105]. In fact, much research has focused on improving the effectiveness of grout in adverse environments associated with the disposal of radioactive waste [106–108]. As discussed in these references, a variety of cement-polymer composites have been investigated as a means of making grouts more compatible with the radioactive and chemical constituents in waste.

(a) (b) (c)

6.3 Examples of composite waste forms using encapsulation in cements where the cement physically surrounds the waste and the radionuclides may be immobilized by being incorporated into the cement phases. The ILW in (a) is compacted ILW solids, in (b) is Magnox (Mg alloy) fuel cladding swarf and in (c) is ceramic fuel zircalloy cladding hulls.

For example, the addition of blast furnace slag to the Saltstone cement[8] being used to solidify Cs-decontaminated salt supernate at the Savannah River Site (SRS), provides a chemical reductant [iron(II)] and a precipitating agent (sulfide) that chemically binds contaminants such as chromium and technetium as insoluble species, thus reducing their tendency to leach from the waste form. Experimentation has shown that leaching of chromium and technetium was effectively reduced to levels that would allow all projected future salt solution compositions to be processed into Saltstone [109]. Long-term lysimeter studies have shown that the addition of slag essentially stopped technetium-99 leaching, although it did not reduce nitrate leaching [109]. Because the SRS Saltstone admixture that is blended with 45% liquid waste is only 10 wt% OPC, 25 wt% fly ash, and 25 wt% slag, it is a geopolymeric cement as the alkali in the salt supernate reacts with the fly ash in geopolymer-like chemical reactions.

The water in the hydrated cement blends may generate H_2 by radiolysis in high radiation fields and require vented canisters [110] when containerized. While this study concentrated on transuranic (TRU) wastes containing ^{238}Pu oxide, which is primarily alpha radiating, the other studies have demonstrated the radiolysis of concrete with ^{60}Co (gamma radiation) and ^{3}H (beta radiating) [111–113].

Recent comprehensive reviews of cement systems for radioactive waste disposal can be found in Pabalan *et al*. [114] and Glasser [115]. Long-term cement durability comparisons have been made using ancient cements, geopolymers, and mortars [116–123], some of which may also serve as natural analogues for geopolymer wasteforms [124, 125].

The cements and grout formulations are too extensive to list as examples. The durability response is complex due to the relative response of encapsulation with some chemical reaction and embedding. Therefore, the durability is usually modeled as a diffusion rate with respect to the element(s) of interest.

Geopolymers

Forming geopolymers is a process that is very similar to cementation. Geopolymers are inorganic ceramic polymers made from aluminosilicates and cross-linked with alkali metal ions [126–128]. During fabrication, a low water content is used (H_2O/M_2O ~ 10–25 wt%) so that an amorphous geopolymer forms instead of crystalline zeolites which would technically form hydroceramic waste forms discussed below. A nominal composition of $4SiO_2 \bullet Al_2O_3 \bullet M_2O$ is used to represent the geopolymer matrix, although

[8]Saltstone contains 5 wt% cement, 25 wt% flyash, 25 wt% blast furnace slag, and 45 wt% salt solution.

the Si:Al ratio varies according to the application from 1 to 3. For cements and concrete-like applications, a ratio of 2:1 is nominally used [129]. The alkali can be Na, K, or Cs. Geopolymers appear to be excellent low temperature binders and environmentally more acceptable than cement waste forms as the starting materials only need to be heated to ~700°C instead of clinkering at 1,400–1,500°C.

Geopolymers and geopolymeric cements, including but not limited to fly ash-based geopolymeric concretes, are ideal for environmental applications, such as the permanent encapsulation of radioactive species [130, 131] and other hazardous wastes [132]. Geopolymers can be used as sealants, capping, barriers, and other structures necessary at containment sites. Pilot-scale demonstrations have been performed in Europe on both mining wastes and uranium mill tailings [133–135]. Geopolymers were investigated for the disposal of radioactive wastes in Europe in the mid to late 1990s [136, 137] and the following applications have more recently been investigated.

- Geopolymers with Si:Al ratios of 1:1 and 2:1 for the stabilization of hazardous Resource Conservation and Recovery Act (RCRA) metals such as Ni, Se, Ba, Hg, Cd, Cr, Pb. A simulant RCRA spike was made that contained the RCRA components at 60× the concentration of the RCRA treatment standards known as the Universal Treatment Standards or UTS limits [138]. The mixture was very acidic (pH < 1). The RCRA simulant was substituted for half of the 10 wt% water in the geopolymer formulation and the geopolymers met the Environment Protection Agency Toxicity Characteristic Leaching Procedure (EPA TCLP) test limits at less than the UTS limits even though the geopolymer contained 60× the UTS concentrations. It is not known whether or not the RCRA components interacted with the geopolymer, i.e. whether this was encapsulation or embedding (Table 6.9).
- Geopolymers derived from metakaolin and alkaline silicate solutions and having nominal Na/Al and Si/Al molar ratios of 1 and 2 were studied at ANSTO for the stabilization of ^{137}Cs and ^{90}Sr [139]. These geopolymers were studied by transmission electron microscopy and found to be amorphous on the ~1 nm scale after curing at 40°C. The Cs inhabited the amorphous phase, whereas Sr was incorporated only partly, being preferentially partitioned to crystalline $SrCO_3$. This study implies that the geopolymer components do interact with some species and not with others, providing both encapsulation and embedding (Table 6.9).
- Special geopolymer formulations, marketed under the name DuraLith, have been patented [140] for stabilization of ^{129}I and ^{99}Tc. Testing [141] showed great promise for retention of technetium with rhenium used as a surrogate for the Tc, but not for iodine.

- Removal of radiolytic H_2 production (and freeze-thaw problems) can be carried out by heating geopolymers at ~300°C without any serious effects on strength or leachability [142].
- Geopolymers have demonstrated excellent fire resistance [142].

Hydroceramics

Hydroceramics are another concrete-type material that is similar to zeolitized rock. It is made by curing a mixture of inorganic waste, calcined clay, vermiculite, and Na_2S, NaOH with water under hydrothermal conditions (60–200°C) to form a matrix containing crystalline zeolites embedded in a sodium aluminosilicate matrix [143]. The solidification process occurs as a result of hydration reactions. The NaOH solution dissolves the metakaolin ($Al_2O_3 \bullet 2SiO_2$) much the same as in geopolymers, but abundant water or hydroxides provide the water to create crystalline silicates instead of an amorphous matrix. The hydroceramic process takes advantage of the sodalite and cancrinite structures in immobilizing oxyanion salts such as nitrate, nitrite, chloride, fluoride, and iodide within the physical cage-like structures of the crystals created.

Hydroceramic waste forms have been shown to be effective on low-activity sodium-bearing waste. The technology is still under investigation with studies focused on optimization of waste pre-treatment (calcination), waste stream-specific optimization of the formulations, and a study of scale-up factors to ensure viability for full-scale operation [143]. In cases where the waste has a high nitrate-nitrite composition, the waste must first be denitrified in some manner, such as calcination, to remove the nitrates and nitrites from the waste. If sodium nitrate-based waste is pre-treated with metakaolin, sucrose, and then calcined, it can be used to make a hydroceramic waste form [143]. Successful waste forms have been achieved with waste loadings of 40–60 wt% waste [144, 145]. Hydroceramic waste forms have been made with Idaho National Laboratory's HLW calcine [146].

Ceramicrete

Phosphate-bonded ceramics, also known as chemically bonded phosphate ceramics, form through the reaction of magnesium oxide with mono-potassium phosphate in water according to the following reaction:

$$MgO + KH_2PO_4 + 5H_2O \rightarrow MgKPO_4 \bullet 6H_2O$$

The reaction product ($MgKPO_4 \bullet 6H_2O$) is Ceramicrete, a rapid-setting phosphate ceramic [147] that contains a considerable amount of bound water. The reaction takes place at room temperature, although there is some heat generation from the reaction, to form a hard, insoluble ceramic. Some

waste components react to form insoluble phosphates, and others are encapsulated in the matrix. The patented technology [148] has been licensed to treat mixed and LLW and is being used for macro-encapsulation and containerization of uranium. In the US, this low temperature waste form has also been investigated for both micro- and macro-encapsulation of radioactive and hazardous waste streams [141].

The waste treatment process includes neutralizing the waste to a pH of 5; adding sodium sulfide, tin chloride, and silver zeolite to precipitate insoluble compounds of Hg and Cr, Tc(Re), and I, respectively; evaporating water to reduce the volume; and adding the binder mix (MgO, KH_2PO_4, $CaSiO_3$). Adding silica as wollastonite ($CaSiO_3$) or fly ash improves the waste form performance [141].

Bitumen

There are several processes for solidifying bitumen with waste streams [6]. The most common bituminization process embeds wastes in molten bitumen and the waste becomes encapsulated when the bitumen cools. The process combines heated bitumen and a waste concentrate, usually in slurry form, in a heated extruder containing screws that mix the bitumen and waste. Water is evaporated to ~0.5% moisture [149]. The final product is a homogeneous mixture of extruded solids and bitumen.

Bituminization has proven to be effective in treating LLW. Radionuclides are effectively controlled and Ojovan *et al*. [150] have recently shown that, as the bitumen ages, it becomes harder and more thermally stable. These assessments of aged bitumen had been made after 12 years in an open (wet) repository. The bitumen was found to age into asphaltenes, saturated hydrocarbons and aromatic hydrocarbons. The asphaltene fraction was found to increase as the bitumen aged and the asphaltene was found to be responsible for retaining the major part of the radioactivity. Thus bitumen was found to be as durable as some glasses. This is in keeping with the much earlier findings of Westsik [151] who showed that bitumen was more durable than cement waste forms with very low fractional release rates of $<10^{-5}$ fraction/day. Bitumen has been used in Europe, Canada, Russia and to a lesser extent in the United States.

6.4.3 Solidification in composites (chemical incorporation and encapsulation)

Composites can be thought of as multi-barrier waste forms. Usually a composite waste form is required to meet a specific waste form criterion, e.g. heat loading, respirable fines, compressive strength, etc. A single-phase or multiphase crystalline ceramic or even a glass can further be encapsulated

in a metal, a glass, or an encapsulant waste form such as cement, geopolymers, hydroceramics, bitumen, etc. The encapsulant phase offers a second level of protection to the release of radionuclides or hazardous components in the waste form as shown in Table 6.10. Composites include many GCMs such as glass bonded sodalites that have already been discussed (on page 214). Composites can also include deteriorated cement waste forms that are remediated by encapsulation (see Table 6.10). A few examples are given below and others are shown in Fig. 6.3.

Metal matrix

In metal matrix waste forms a metal is used as the encapsulant for either glass or crystalline materials in which the radionuclides or waste species are already atomically bonded. The advantages of this type of encapsulation include (1) improved thermal conductivity of the waste package, (2) potentially decreased leach rates of radionuclides because of the metal matrix encapsulation, (3) improved mechanical strength and decreased dispersability on impact, and (4) improved radiation protection during handling [152, 153]. The encapsulation of waste forms in metal matrices was pursued in the US and developed full scale in PAMELA, which was a joint Belgian–German project located in Belgium.

Vitromelt is a composite waste form in which glass beads (0.5 cm) are embedded in a metal matrix (usually a Pb alloy) [154–156]. For example, waste immobilized in calcium silicate pellets was encapsulated in a lead matrix. In one variation of the commercial PAMELA vitromelt process, phosphate glass beads containing HLW were produced by passing molten glass through nozzles. The beads were subsequently fed into a container and infiltrated with molten lead alloy to produce a composite waste form ('vitromet'). The beads, with a diameter of 0.5 cm, occupy up to 66% of the total volume. Increased thermal conductivity of the waste form leading to lower waste temperatures is one of the most important advantages of this product.

In studies related to vitromelts, immobilized waste pellets have been coated with pyrolytic graphite, before encapsulating in a metal matrix, in order to improve the leach resistance. Application of other coatings has also been reported, including alumina, titania, silica, silicon carbide, chromium silicide, and chromium oxide, together with a variety of metals including Ni, Fe, and Mo. Dual coatings of pyrolytic graphite and alumina have also been reported. Metal matrices have included Pb-based alloys (e.g., Pb–Sb, Al, Sn), Al-based alloys (e.g., Al–Si, Cu, Ti), and Cu. Particles can be coated by conventional ceramics (e.g., Al_2O_3, TiO_2, or SiO_2) or by carbon products (e.g., PyC, Cr_7C_3, or SiC), glass (borosilicate or aluminosilicate), or metals (e.g., Ni, Si, or Fe) before being encapsulated). Uncoated, sintered supercalcine pellets have been encapsulated in vacuum-cast Al-12Si and

glass-coated, sintered supercalcine pellets encapsulated in vacuum-cast Al-12Si. Supercalcine pellets have also been PyC/Al_2O_3 coated before encapsulation in gravity-sintered Cu.

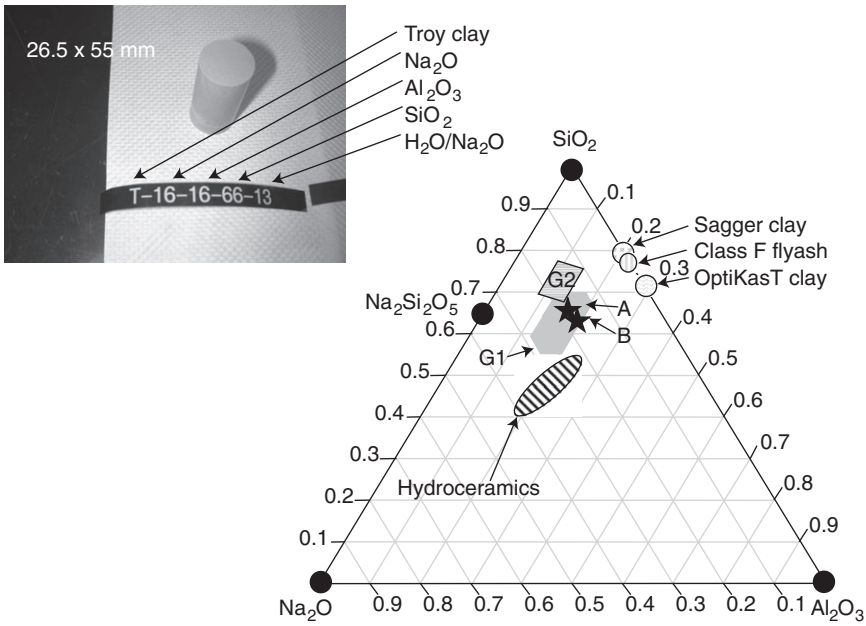
Cermets are related composite waste forms in which radionuclides in the form of small oxide or silicate particles +1 mm in size are dispersed in a metal matrix [152]. The unique aspects of the waste form are the very fine scale on which the radionuclide-containing phases are dispersed, the fact that the alloy is primarily composed of hydrogen reducible metals which are already in the waste, the high thermal conductivity, and reduced leach rates due to the alloy encapsulation. Developmental work on cermet was performed using simulated wastes, radionuclide-containing simulated wastes, West Valley acid THOREX wastes, and SRS HLW sludge and unneutralized SRS wastes. Waste loadings of up to around 30% have been reported. The addition of elements in excess of stoichiometric requirements is used to guarantee the formation of specific ceramic phases, e.g. excess Al and Si to ensure the formation of pollucite.

Ceramic/mineral waste forms created using FBSR in a geopolymer or cement

Use of the FBSR process to produce a highly leach resistant mineralized waste form from Hanford low activity waste (LAW) has been investigated since 2001 (see page 217). Initial studies focused on producing and testing the granular mineral product created by processing high sodium waste feeds with clays at ~720°C to produce nepheline ($NaAlSiO_4$) and nepheline-based minerals such as the sodalites to host I, F, Cl, and nosean to host sulfate and sulfide. Numerous studies (74–80) have shown that it is possible to produce a mineral waste form that effectively immobilizes both radionuclides and hazardous constituents.

To be accepted for near-surface disposal, the waste form is required to meet an acceptance criterion for compressive strength of 500 psi. This requirement is derived from a Nuclear Regulatory Commission Branch Technical Position on low level waste (LLW) forms in the US, which somewhat arbitrarily specifies 500 psi to preclude subsidence in the waste disposal system. It is also noted that a monolithic waste form reduces the impact to human health for the intruder scenario in the waste site performance assessments. While a monolith is desirable, there are other means by which this requirement can be met, e.g. waste stabilization in high integrity containers (HICs).

In 2005–2006 the Savannah River National Laboratory (SRNL) performed a monolith feasibility study for granular FBSR product [157]. Monoliths were made out of ordinary Portland cement (OPC) at 80–87 wt% FBSR loading, out of ceramicrete (a blend of MgO and monopotassium



6.4 Formulation region for geopolymers compared to hydroceramics in the Na_2O-SiO_2-Al_2O_3 (mol%) ternary. Note that the fourth dimension is water content and not shown on the ternary mol% diagram. The geopolymer region labeled as G1 is the target range. Optimum formulations are designated as A and B and a 1″ × 2″ cylindrical monolith made with composition A is shown in the photograph.

phosphate (KH_2PO_4)) at an FBSR loading of 35.7 wt%, and out of hydroceramics (aluminosilicate zeolite phases formed from metakaolin plus NaOH) at FBSR loadings of 50–80 wt%. The hydroceramics had the best durability as they had a similar chemical makeup to the FBSR product (see Fig. 6.4) but the hydroceramics required hydrothermal processing. Therefore, geopolymers were used to bind the granular mineral waste form due to the similarity of the chemical makeup (see Fig. 6.4) to the FBSR product and the fact that the geopolymers did not require hydrothermal processing. Up to 70 wt% granular product was stabilized in the geopolymer. The granular mineral stabilized geopolymer was shown to be more durable than the granular product alone [158].

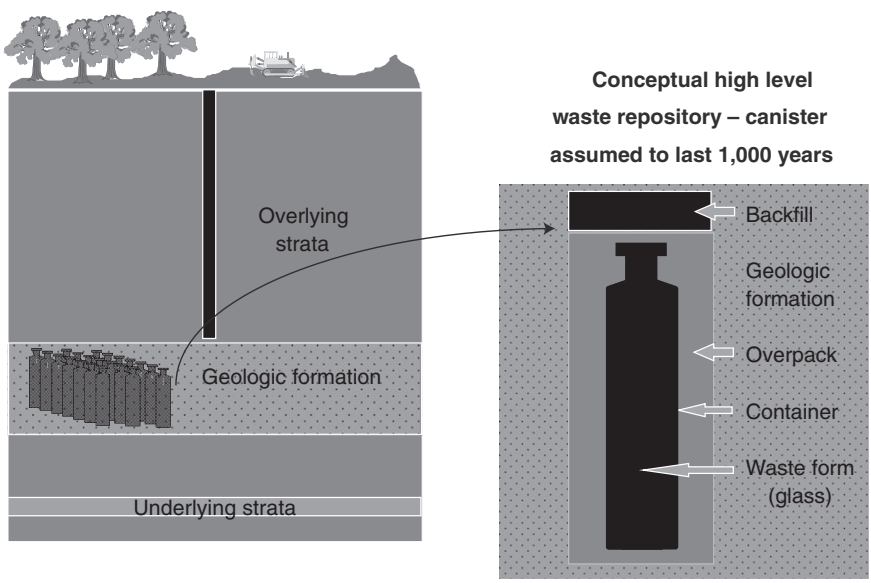
6.5 Waste forms, waste packages, and the geological environment

The long-term behavior of a waste disposal facility is a function of the entire disposal system, including the waste form, engineered barriers, and

surrounding environment. In order to assess the ability of a given disposal concept to meet regulatory requirements, it is necessary to consider the influence of each of these system components on short- and long-term performance. This is accomplished through the performance assessment (PA) process. For HLW, many countries are proposing long storage life for the canistered glass waste forms during geological repository siting and preparation. During that time, a great many of the radionuclides will decay leaving the long-lived radionuclides as the primary sources that need be considered in a PA.

Figure 6.5 is a schematic of a generic high level waste repository. It shows the relative role of the waste form, the role of the multiple barriers (canisters, containers, overpacks, and casks) in the waste disposal system. It is the multi-barrier concept – a barrier within a barrier within a barrier as discussed in Chapter 1. Ultimately the role of the repository or disposal environment is to isolate the waste from the biosphere until all the barriers have failed at which time almost all of the radionuclides will have decayed.

While the waste form is the source term and should be as durable as reasonably possible, multiple barriers must corrode before the waste form will be exposed to groundwater. As a result of the research programs over the past several decades, there is now an extensive database and substantial understanding of the behavior of nuclear waste glasses in a variety of disposal environments [159]. The present challenge is to model glass behavior



6.5 A generic HLW waste disposal system.

in the near-field of specific geologic repository environments and to develop a fundamental understanding of the long-term corrosion rate [160].

6.6 Recent advances in waste form processing

Historically, the crystallization of vitreous waste forms has always been regarded as undesirable, as it has the potential to alter the composition (and hence, durability) of the remaining continuous glass phase, which would (eventually) come into contact with water. However, there has been a recent trend toward higher crystallinity in ostensibly vitreous waste forms so that they are more correctly termed GCMs. This is particularly apparent in the development of hosts for more difficult waste or where acceptable durability can be demonstrated even where significant quantities of crystals (arising from higher waste loadings) are present. Acceptable durability will result if the active species are locked into the crystal phases that are encapsulated in a durable, low-activity glass matrix. The GCM option is being considered in many countries, including Australia, France, Russia, South Korea, the UK, and the US. The processing, compositions, phase assemblages, and microstructures of GCMs may be tailored to achieve the necessary material properties.

Joule heated melters are relatively intolerant of crystal growth in the melt which causes slag formation [161]. Recently, Sellafield has shown the ability to go to 38 wt% waste loading [162] from 25 wt% waste loading [163] by allowing spinel formation in the melt, but the Sellafield melter is induction heated not a JHM design. However, 1–2% crystallization of spinels is planned for Hanford's HLW AJHM and it is anticipated that the spinel crystals will stay buoyant from the melt pool agitation afforded by the bubblers [164, 165]. This strategy can be made to work unless during long maintenance outages when the melter is idled, the crystals grow larger than the size that the agitation can sustain. Otherwise, the melt pool will have to be diluted with components that dilute the spinel-forming tendencies because JHMs and AJHMs cannot be drained without causing damage to the electrodes. In addition, the spinels that form cannot be redissolved into the melt except at >1,400°C, which is a temperature that cannot be achieved with the Inconel® electrodes in the AJHM. Therefore, cold crucible induction-heated melters (CCIM), which are already being pursued in Russia, France, and the US, will have to be substituted as an alternative to JHM and AJHM melter technology. The major advantages of CCIM over JHM/AJHM are higher productivity, higher temperatures, longer lifetime, smaller dimensions, can be drained as the heating is external, and can be stirred which allows higher waste loadings while maintaining the same product quality. Thus CCIM is robust in terms of producing GCMs and mineral waste forms by a melt and controlled crystallization route.

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Advances in the techniques to measure and quantify how and where radionuclides are bonded in glasses and glass ceramics will enable GCMs to be tailored to sequester the desired radionuclides in the ceramics phases and either minimize or prevent the radionuclides from migrating to the glassy encapsulating phase. This will allow the crystalline and glass structures based on MRO and LRO to be used to model glass and GCM behavior and properties.

Mineral waste forms will advance using novel processing techniques like templating. Hybrid waste forms, e.g. glass-ceramics instead of glass vs. ceramic, geopolymeric cements combining geopolymers and cement, or methods that combine thermal treatment (calcining, FBSR) with encapsulation in geopolymers or cements will provide double barrier composites for troublesome waste species.

6.7 Radiation damage in glasses and ceramics

No discussion of the relative attributes of glass, glass-ceramics, and ceramic/mineral waste forms is complete without a discussion of the relative radiation stability of the various waste forms. The effects of radiation damage due to self-irradiation of various waste forms have been studied for decades and a few of the more recent reviews of this field are given in Section 6.9.2.

Radiation damage is not only important because it impacts the stability of the wasteform but if the material swells or is otherwise degraded by self-irradiation, this impacts the long-term durability of the waste form (see discussion in Section 6.8). While glasses undergo radiation damage that can cause density variations, careful durability experiments have shown that the radiation damage does not affect the dissolution rate as studied by single pass flow through (SPFT) and Soxhlet durability tests. Many pertinent examples are given in [4].

In ceramic/mineral waste forms, the crystalline structure may become amorphous, a phenomenon observed in nature and known as metamictization (the formation of amorphous metamict phases). This phase change, from a crystalline structure to a partially or completely amorphous structure, is accompanied by macroscopic swelling of the structure. An interesting study by Weber *et al.* [166] has shown that the Synroc crystalline phases (zirconolite and perovskite) are susceptible to macroscopic swelling at high dosages (Fig. 6.6). Moreover, the swelling in the titanium-based Synroc phases is greater than the swelling in the silicate-based supercalcine ceramics at lower dosages which in turn is greater than the swelling in glass ceramics (Fig. 6.6). Additional references regarding the swelling of individual mineral/ceramic phases can be found in the references in the annotated Tables 6.6 and 6.8.

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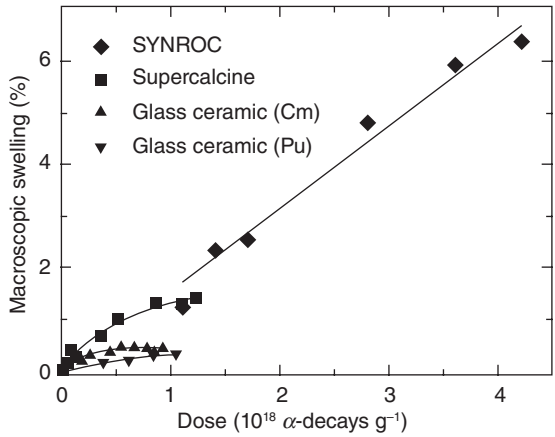


6.6 Macroscopic swelling in Pu-doped Synroc, Cm-doped supercalcine, and Cm-doped and Pu-doped glass-ceramics. (Reprinted with permission from [166].

6.8 Leach testing and its role in the waste acceptance process

The most important requirement for a waste form is its chemical durability, expressed as a dissolution rate. It should be noted that for some radionuclides, solubility limits the dissolution rate while others are completely soluble, e.g. ^{99}Tc, ^{129}I, or ^{135}Cs. These soluble radionuclides are released at the maximum forward rate of dissolution. For the production of durable nuclear waste forms, it is desirable for the waste forms to be highly insoluble in the long term to minimize release to the environment, i.e. to have the slowest forward dissolution rate possible. Since no 'durability test' can be carried out on these geologic timescales, dual approaches are taken:

1. Durability test parameters such as surface area (SA), time (t), temperature (T), or a combination such as (SA)•(t) are used to 'accelerate' dissolution as long as the acceleration parameter(s) used does not change the dissolution mechanism. To ensure that the mechanism is not 'altered' by the acceleration modes of the experiments, natural analogs are usually tested simultaneously.
2. Models are used to predict waste form dissolution from parameters that can be measured such as the activation energy of dissolution, forward rate of dissolution, and from an understanding of the dissolution mechanisms. Predictive and/or transport models for waste form performance on extended time scales (1,000–1,000,000 years) has led to various thermodynamic and kinetic models (see [160], [187]).

Thus, there are no 'waste form-specific' durability tests, but a suite of tests that must be performed to understand the leaching mechanism(s) of a waste form and to derive the parameters necessary for the particular predictive or transport model(s) being applied.

In order to determine if a particular waste form is acceptable, it must be demonstrated that the waste form performance in the disposal system is adequate. Such evaluations in the US are known as total system performance assessments (TSPA) for HLW and performance assessments (PA) for shallow land disposal of immobilized LAW known as ILAW in the integrated disposal facility (IDF). The TSPA or PA includes all of the testing and performance modeling that has been gathered on the waste form and the TSPA is intended to provide a technical basis that a waste form is acceptable for deep geological disposal.

For HLW in many countries the geological disposal sites have not been determined while wasteform producers have already made many canisters of vitrified waste (see Table 6.1). Due to the mismatch in timing between the need to stabilize HLW and when a geological repository will be chosen and ready to receive the wasteforms, the US devised a strategy to addresses vitrified waste acceptance based on production control. Production control is intended to determine how the production of a waste form material affects (or controls) its performance and identify the ranges for processing variables that result in an acceptable waste form. The primary role of most of the waste acceptance product specifications (WAPS) developed in the US for vitrified HLW waste forms verify that the properties of a specific waste form product are consistent with the existing regulations and thus will be acceptable for disposal, either by direct measurement or through process control.

Therefore, waste acceptance testing is, for the most part, focused on comparing a specific waste form product to the range of waste forms that are (1) considered to have acceptable performance based on performance modeling and (2) produced within the production control limits. What will be acceptable with respect to waste form performance and processability will depend on the disposal site and engineered system and cannot be completely quantified at the time the waste form is made. The range of acceptable waste form compositions will depend on the required performance [167].

While the predicted long-term durability of a waste form is a necessity for its 'qualification for shallow land burial' or 'deep geologic disposal', there is also a need for short-term testing that can be related to acceptable performance by the following linking relationships [168]:

$$\text{process control} \leftrightarrow \text{composition control} \leftrightarrow \text{dissolution rate control} \leftrightarrow \text{performance control} \leftrightarrow \text{acceptable performance}.$$

This approach allows a waste form producer to ensure that the waste form that they are producing on a tonnage per year basis will be acceptable to long-term performance instead of having to test each and every canister or form produced. For HLW glass (alkali borosilicate glass) in the US, the manner in which this was done is given below in a brief stepwise fashion and explained in more detail in Refs [11, 169–173]:

1. Develop an acceptable waste form durability based on HLW performance modeling (fractional dissolution rates between 10^{-4} to 10^{-6} parts per year (i.e., the glass waste form would take 10,000 to 1,000,000 years to totally dissolve [174]).
2. The middle of the range determined by HLW performance modeling was adopted as the waste form specification; if the long-term fractional dissolution rate of a wasteform was $\leq 10^{-5}$ parts per year for the most soluble and long-lived radionuclides, then borosilicate glass would provide acceptable performance for any repository site or concept.
3. Develop an understanding of the glass durability mechanisms from a combination of the test protocols (ASTM C1220 which was previously known as MCC-1, ASTM C1285 which is known as the Product Consistency Test (PCT) [175, 176], ASTM C1662 which is the SPFT test, and ASTM C1663 which is the Vapor Hydration Test or VHT).
4. Develop a glass standard, the Environmental Assessment (EA) glass [177, 178] that bounded the upper release rate found to be acceptable from the HLW repository modeling from step 1 above.
5. Generate databases for modeling the maximum radioactive release rate(s) by relating the release of ^{99}Tc, ^{129}I, and ^{135}Cs to the release of non-radioactive species such as Na, Li, and B which leach at the same rate (congruently); this is part of the ASTM C1285 (PCT) test protocol.
6. Develop a short-term test and process control strategy for ensuring that every glass produced has a dissolution rate less than that of the EA glass at the l95% confidence level based on Na, Li, B which in turn ensures acceptable performance control.
7. Continue to qualify that the radionuclide response of production glasses verify that production glass radionuclide releases are consistent with the releases predicted by Na, Li, and B.

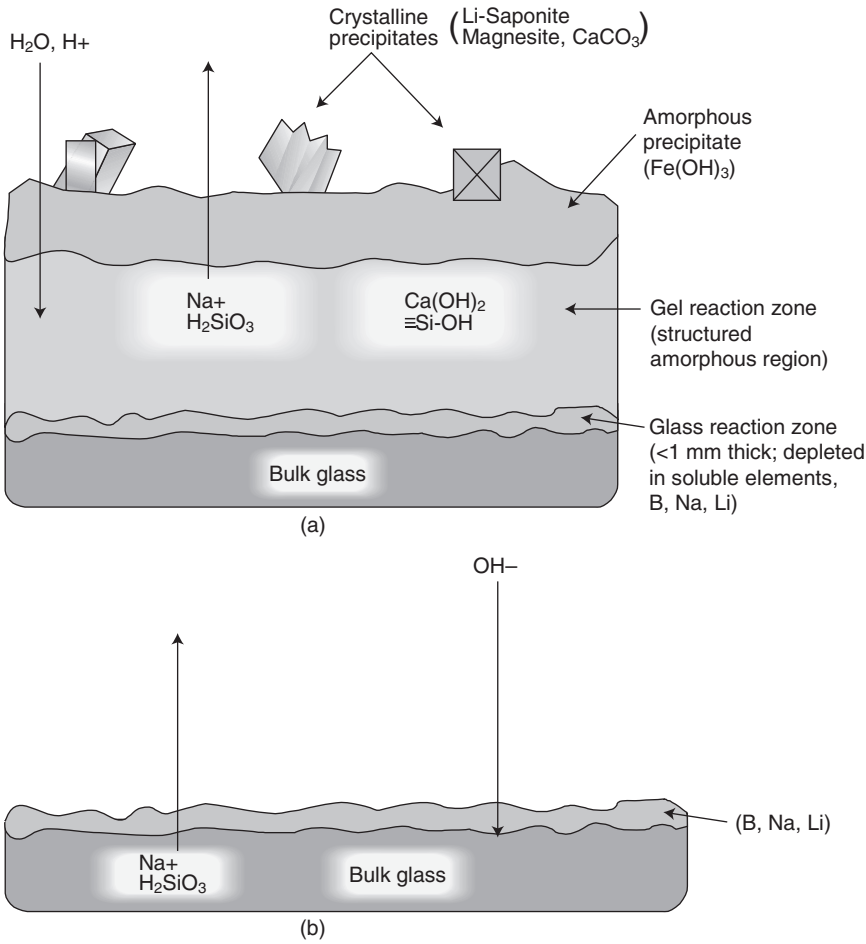
Therefore, a suite of the existing durability tests (those for affinity control, solubility control, and/or diffusion control) must be performed on a waste form to determine the mechanisms, and determine the parameters necessary for the mechanistic model(s) being developed, e.g. the transition state theory (TST) models used in the TSPA for HLW geological disposal or the PAs for shallow land burial. Different durability tests are used for a diffusion model, for example for cement. However, one cannot apply a glass

standard that leaches by an affinity limited mechanism to cement that leaches by diffusion, nor can one apply a borosilicate glass standard to non-borosilicate-type glasses since it is not known whether the radionuclides in non-borosilicate glasses leach by the same degradation mechanism and whether the leaching of Na, Li, and B remain congruent with the leaching of the radionuclides. In these cases, new standards need to be developed and qualified and the leaching mechanisms understood.

For glasses, the advances in the measurement of medium range order (MRO) in glass waste forms has led to the understanding that the molecular structure and composition of a glass, like the molecular structure and composition of minerals, controls the waste form durability by establishing the distribution of ion exchange sites, hydrolysis sites, and the access of water to those sites. During the early stages of glass dissolution, a 'gel' layer resembling a membrane forms through which ions exchange between the glass and the leachant (Fig. 6.7). The hydrated gel layer exhibits acid/base properties which are manifested as the pH dependence of the thickness and nature of the gel layer. Advances in the understanding of the dissolution mechanisms of borosilicate glasses proposed for nuclear waste solidification were extensively studied in the 1980s–1990s [22, 179–186] and such mechanisms are still being studied [160, 187–190]. At least four operative mechanisms have been shown to control the overall glass durability as shown in Fig. 6.7. These four mechanisms are ion exchange, matrix dissolution, accelerated matrix dissolution, and surface layer formation (possibly of a protective or passivating nature).

One can bound or model the shorter term durability of a glass using kinetic or thermodynamic models to describe the impacts of ion exchange and matrix dissolution or hydrolysis by examining either time-temperature data (Fig. 6.8) or release vs time or accelerated release, expressed as SA/V•time (Fig. 6.9), but these underlying mechanisms become modified if surface layers form and/or if, over very long periods of time, the gel layer ages *in situ* into clay or zeolite minerals or the leachate becomes saturated with respect to a clay or zeolite phase. If zeolite mineral assemblages (higher pH and Al^{3+} rich glasses) form, the dissolution rate increases (Fig. 6.9) which is undesirable for long-term performance of glass in the environment.

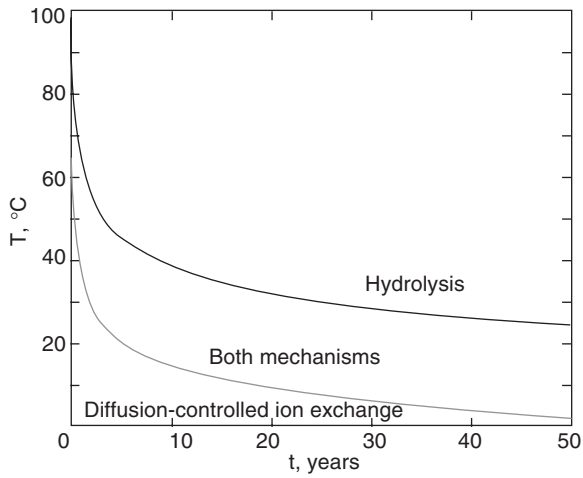
The current theories of glass dissolution [159] suggest that all glasses typically undergo an initial rapid rate of dissolution denoted as the 'forward rate' (Figs 6.8 and 6.9). However, as the contact time between the glass and the leachant lengthens, some glasses come to 'steady state' equilibrium and corrode at a 'steady state' rate, while other glasses undergo a disequilibrium reaction with the leachant solution that causes a sudden change in the solution pH or the silica activity in solution [191]. The 'return to the forward rate' (Fig. 6.9) after achieving 'steady state' dissolution is undesirable as it



6.7 (a) Schematic diagram of glass dissolution mechanisms (ion exchange and matrix dissolution) in aqueous solution, coupled with both hydrated amorphous surface layer formation and crystallization/precipitation from solution [179, 402]. (b) Schematic diagram of the glass dissolution mechanism known as 'accelerated matrix dissolution.' In this mechanism, the excess strong base in the leachate released by the ion exchange mechanisms attacks the glass surface layers, including the gel layer, and makes the glass appear to have little or no surface layer.

can cause a glass to return to the rapid dissolution characteristic of initial dissolution.

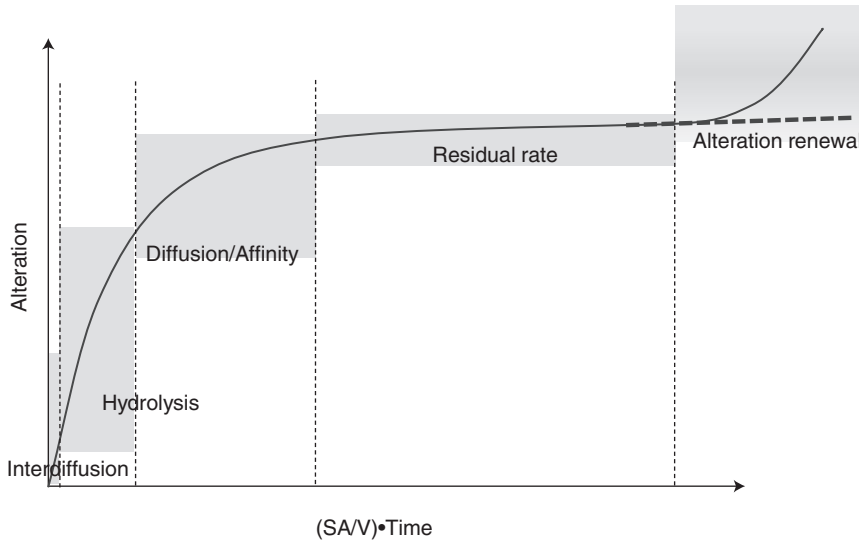
The initial rate is often referred to as Stage I dissolution in the US literature, but it encompasses zones where multiple mechanisms are operative including regimes that are interdiffusion controlled, hydrolysis controlled, and a rate drop that is diffusion or affinity controlled [159]. The 'steady



6.8 A temperature–time plot of the incongruent corrosion mechanisms exhibited by British Magnox waste glass in deionized water, showing that corrosion in deionized water at a constant temperature begins immediately with an instantaneous surface dissolution followed by a diffusion controlled ion exchange phase. As corrosion progresses, the impact of hydrolysis becomes significant with comparable contributions from both ion exchange and hydrolytic reactions. Finally, glass corrosion in deionized water is fully controlled by hydrolysis [36].

state' rate (also known as the residual or final rate) that signals the end of the alteration phase and/or a pseudo-equilibrium between the alteration and re-condensation reactions [159, 192] is known as Stage II dissolution, and the return to a forward rate (or resumption of alteration) is known as Stage III dissolution. Diffusion controlled dissolution of network modifiers and/or radionuclides during Stage I and Stage II normally follow a mathematical function related to the square root of the test duration as observed in many burial studies [190], while other radionuclides are solubility limited, entrapped in the gel layer, or complexed in secondary alteration phases that form from the leachate solution.

A reaction zone is formed as the leached layer solution interface progresses into the glass (Fig. 6.7a). The front of the reaction zone represents the region where the glass surface sites interact with the ions in solution [193]. The top of the gel reaction zone represents the leached layer–glass interface where a counter-ion exchange occurs [193]. The glass dissolution rate is modified by the formation of the hydrated amorphous gel layers and/or secondary precipitates, e.g., metal hydroxo and/or metal silicate complexes that have reached saturation in the leachate and can precipitate on the surface of the gel layer [22, 179, 181, 182, 194, 195]. These 'back reactions' have been attributed to formation of silanol bonds as surface



6.9 Parabolic behavior of the diffusion profile of soluble species out of a waste glass through an increasingly thick surface layer [159]. Acceleration of glass durability tests using glass surface area (SA), leachant volume (V), and time. Acceleration appears to follow parabolic diffusion kinetics until SA/V is ~20,000 m^{-1}, when the glass dissolution mechanism appears to change reverting to a rate similar to the forward rate but likely controlled by precipitation of secondary phases.

adsorption sites which were modified by changes in solubility of the species in solution and surface (zeta potential) considerations [22, 196].

The gel layer may, under certain conditions, act as a selective membrane [194, 197] or as a protective/passivating layer [22, 159, 180–182, 184–186, 192, 198]. The slowing of glass dissolution to a steady state rate by solution saturation (affinity) of glass matrix elements or reaction through a surface layer has been referred to as Stage II dissolution including residual rate dissolution, steady state dissolution, or the final dissolution rate. Recent mechanistic modeling of glass durability, including the slowing of the dissolution rate due to affinity and/or surface layer effects, was first modeled by Grambow and Muller [199] and is referred to as the GM2001 model. The GM2001 model combines the effect of glass hydration by water diffusion with ion exchange and affinity-controlled glass network corrosion (Figs 6.8 and 6.9). The slowing of dissolution due to the effect of a growing surface gel layer is represented by a mass transfer resistance for silica by this layer. At the interface between the glass and the gel layer, a different 'gel layer' is assumed to be hydrated glass that allows diffusion of H_2O in and boron and alkali atoms out of the glass (similar to Fig. 6.7). A 2003 modification

of the GM2001 model, known as the GM2003 model [159], treats silica dissolution and silica diffusion through the gel separately from water diffusion, and boundary conditions are specified at the gel/diffusion layer and the gel/solution interfaces. Recently, the GRAAL (glass reactivity with allowance for the alteration layer) model [187, 189] has been proposed, which is dependent on the composition and the passivating nature of the gel layer, called the passivating reactive interphase (PRI). The leached layer has been found experimentally to be zoned (5–7 zones) and the GRAAL model assigns various mechanisms to different zones within the PRI.

The resumption of alteration (Stage III) causes the long-term dissolution rate to reaccelerate to a rate that is similar to the initial forward dissolution rate for some glasses. This unexpected and poorly understood return to the forward dissolution rate has been shown to be related to the formation of the Al^{3+}-rich zeolite, analcime, and/or other calcium silicate phases. Moreover, the presence of Al^{3+} and Fe^{3+} in the HLW glass, in the leached layer, and in the leachant has been shown to influence whether a glass maintains Stage II dissolution or reverts to the forward rate of dissolution, e.g., Stage III dissolution. Van Iseghem and Grambow [191] demonstrated that an Al^{3+}-rich zeolite (analcime) formed on certain glasses during dissolution but not on others. Van Iseghem and Grambow also demonstrated that a change in solution pH accompanied the return to the apparent forward rate when analcime formed. Likewise, Inagaki *et al*. [200] demonstrated that solution pH and solution concentrations of Na and K were also involved in the formation of undesirable analcime versus Na-bedellite (a smectite clay). Other zeolites and smectite clays that are rich in Fe^{3+} compared to Al^{3+} do not appear to accelerate glass corrosion [191, 201, 202].

Since many long-term durability models are still being refined and an international study group [203] is actively working on a refined understanding of the PRI, a variety of leaching tests are being used to facilitate an integrated understanding of these stages of durability.

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Assessing and modelling the performance of nuclear waste and associated packages for long-term management

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Abstract: Examples of analytical approaches and methodologies for modelling the behaviour of waste forms and waste package metals in long-term management of spent nuclear fuel (SNF) and high level waste (HLW) are presented. Two cases, long-term geological disposal and interim extended dry storage, are considered. The integrity of the waste package (or canister) that serves as a barrier is dependent upon the performance of construction metals. Corrosion degradation modes of the construction metals are evaluated. The waste behaviour during SNF degradation is also evaluated. In each mode of corrosion or degradation, the associated risk insights are discussed in the system performance of disposal or storage.

Key words: assessment and modelling, nuclear waste form, nuclear waste package, storage, disposal.

7.1 Introduction

This chapter presents example analytic approaches and methodologies for modelling the behaviour of waste forms and different metals used in packaging spent nuclear fuel (SNF) and high level waste (HLW). The long-term behaviour of waste forms and different metals used for packaging SNF and HLW are important attributes in assessing safety and security associated with nuclear waste management. This behaviour is a core component in determining radionuclide source-term and/or criticality control, used in assessing radionuclide release to the human environment. The assessments and modelling of the long-term behaviour of the waste form and different metals are further complicated by a variety of environmental conditions, including natural and human-induced external hazards. This is especially true when the purported waste management time is very long, e.g., several thousand years or beyond.

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The approaches and methodologies presented in this chapter also cover model uncertainties that affect assessment of public health and safety. The content of this work is considered by the US Nuclear Regulatory Commission (NRC) which assesses cases for management of SNF and HLW in the US. The NRC prepares risk and performance insights. Information that the NRC obtained from the past activities in the management of SNF and HLW in the US, along with relevant information from different international programs, is included.

In the US, the long-term management of SNF and HLW is considered for *geological disposal* for thousands of years and beyond, and interim *extended dry storage* of SNF for up to 300 years. In both management cases, materials performance issues related to waste form and corrosion of different construction metals are considered, i.e., container metal in waste packages used for geological disposal, and canister construction metal for extended dry storage. The following four topics related to the waste-form dissolution and corrosion are addressed in this chapter: long-term integrity of passive film, slow general corrosion, and localized corrosion of different metals; stress corrosion cracking (SCC) of carbon steel and stainless steel; SNF degradation; and cladding performance. The discussion of each topic addresses how it applies to the two management cases, as appropriate. Broader performance issues of waste form and different metals under the two management cases are also discussed. Finally, risk insights are addressed with respect to performance (or risk) assessment for the disposal or storage system. Both management cases incorporate laboratory data, analytical models, archaeological (for disposal) and/or industrial (for extended dry storage and disposal). Some similar classes of metals are used in both cases for different purposes. For example, stainless steels are primarily used for extended storage, but may also be considered for disposal. On the other hand, carbon steel is mainly applied in disposal.

7.2 Background

For each topic in the performance evaluation of waste form and waste package (or canister), the associated risk in the disposal or extended storage system needs to be considered. Three risk-related questions are addressed: (i) what can go wrong?, (ii) how likely is it?, and (iii) what are the consequences? Various time-dependent or one-time behaviours of waste form and waste package are assessed with respect to these three questions. To answer the first question, features, events and processes (FEP) for the geological disposal options (Nuclear Energy Agency, 1997), or equivalent (e.g., NRC, 2007; Dasgupta *et al.*, 2002) are identified. Regarding the second question, the identified FEPs or their equivalents are evaluated with respect to given system designs considering normal conditions and accident conditions

from man-made and natural hazards. For extended dry storage, monitoring, inspection and remediation will reduce the safety significance (likelihood or probability) of some FEPs. Once the likelihood or probability of a FEP, or its equivalent, exceeds a threshold value, its consequence may be assessed in terms of confinement failure, radionuclide release, nuclear subcriticality and radiation shielding, or other performance objectives of the total system or subsystems, thus addressing the third question. Implementation of the answers to these three questions are iterative in nature with modifications of design details, until risk assessment or design performance objectives are met. This iterative process also allows early identification of risk-significant issues related to different designs.

Based on the iterative process, the following FEPs associated with the behaviour of waste package or storage canister construction metals are considered significant in the two management cases.

- Long-term integrity of protective passive film for corrosion-resistant metals such as nickel-based alloys, titanium alloys or stainless steel. This allows low general corrosion rates for these metals, keeping the waste package or storage canister intact for a long time.
- Low oxygen or sulphur ion concentration in the reducing aqueous environment for corrosion-allowance metals such as copper or carbon steel. This also allows low general corrosion rates for these metals, keeping the waste package or storage canister intact for a long time.
- Low susceptibility to localized corrosion. Low ratios of chloride to nitrate ion concentrations prevent localized corrosion in nickel-based alloys and stainless steel. Low fluoride ion concentration prevents fast titanium dissolution without loss of adherent protective passive metal-oxide film. Carbon steel susceptible to pitting in the reducing environment is minimal.
- Low susceptibility to SCC and/or hydrogen-induced cracking. The magnitude of residual stress or concentrations of chemical species such as carbonate ions in solution or salt deposits determine the susceptibility of a metal or alloys to SCC and/or hydrogen-induced cracking.
- Low initial manufacturing defects. This allows minimum early mechanical failure of waste package and canister due to manufacturing defects, for all metals considered.

Similarly, the following FEPs related to the waste form are potentially important for containing the SNF or HLW in the two management cases.

- Conditions affecting dissolution, solubility of actinides. Environmental conditions such as reducing aqueous groundwater result in very low dissolution rates of fission products and low solubility of actinides in SNF dissolution. This limits the radionuclide release into the biosphere.

Similarly, near-neutral pH of the aqueous environment also results in low dissolution rates of fission products and solubility limits for actinides in HLW glass dissolution (BSC, 2004).

- Conditions affecting performance of cladding. Conditions such as low hydrogen absorption, temperature, and residual stress in cladding minimize hydrogen-induced cladding failure.
- The high burnup of SNF encased by the cladding may increase or decrease the radionuclide release fraction, affecting radionuclide release in air.

In the following sections, selected specific subtopics from the above list are discussed in depth.

7.3 Corrosion of relevant metals and alloys in mild and near-neutral pH environments

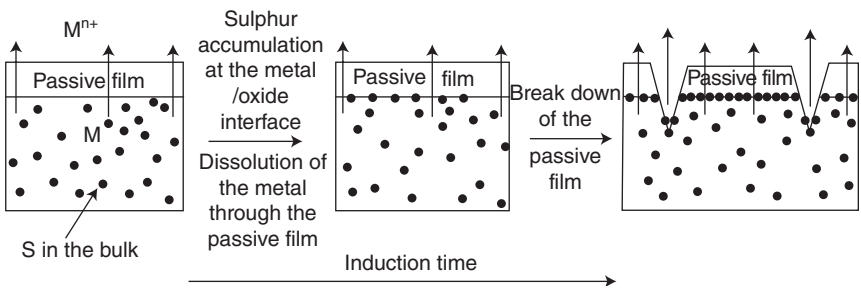
This section presents the general corrosion behaviour of passive and non-passive metal and alloys in mild and near-neutral pH environments such as tuff or granite hostrock sets. Both tuff and granite will have reducing groundwater, whereas tuff can also have oxidizing groundwater. Over a long period of geological time (e.g., several thousand years), a container of a finite thickness is expected to fail even at very slow general corrosion rates. Under more aggressive aqueous environmental conditions that may evolve during the disposal, localized corrosion (e.g., crevice corrosion or pitting corrosion) may occur, decreasing the container lifetime in a very short time (e.g., months to years). The first case to be discussed is long-term integrity of a protective passive film in nickel-based alloy, resulting in very low general corrosion rates. The second case is corrosion-allowance, carbon steel in reducing aqueous environments resulting in very low general corrosion rates. These two cases are discussed further regarding susceptibility to localized corrosion. Finally, risk insights of the general corrosion and localized corrosion in a disposal system are discussed with respect to this type of container with low general corrosion rate or possible susceptibility to localized corrosion.

7.3.1 Long-term integrity of the passive film of nickel-based alloys

In the absence of severe localized corrosion conditions, nickel-based alloys containing chromium are protected against fast corrosion by a chromium-rich oxide adherent film commonly known as a ‘passive film’ at the exposed surface. Typical examples are the thin, adherent passive oxide films observed on sample surfaces after short-term polarization tests and long-term

immersion tests (Orme, 2005; NWTRB, 2002). Film thicknesses were in the range of a few nanometers (10^{-9} meters, nm, 3.9×10^{-10} inch) and tended to be rich in chromium (III) oxides (Cr_2O_3 and/or $NiCr_2O_4$). A thick outer layer was also observed on top of the inner chromium-rich oxide layer. The outer layer was typically porous and consisted mostly of nickel oxide and the oxides of some other alloying elements. The chromium-rich oxide is considered to protect the bare metal against rapid corrosion in the long term, i.e. geological timeframes. A cross-sectional view of the passive film formed on the surface of an annealed nickel-based alloy is presented in Plate II (between pages 448 and 449). It is important to understand whether or not the passive layer persists for a long period of time (Ahn *et al.*, 2008a). A number of issues have been studied to determine whether the protective layer remains stable in the long term. For example, if the protective layer grows continuously, the stress may build up at the interface of the bare metal and the protective layer, and the protective layer may spall off. However, the subsequently exposed bare metal would repassivate. Certain metalloids such as sulphur may be segregated at the interface during the anodic dissolution of the bare metal surface. A potential mechanism of the breakdown of the passive film induced by enrichment of sulphur at the metal–passive film interface is presented in Fig. 7.1 (Marcus, 1995). When the surface concentration of the segregated sulphur exceeds a critical value, the protective layer will become unstable. The bare metal exposed as a result of the unstable protective layer may repassivate after dissolution of the accumulated sulphur layer. Other impurity elements such as silicon in the alloys or solutions may also affect the long-term stability of the protective layer. Microbially-influenced corrosion may also destabilize the protective layer. However, the bare metal surface formed after the destabilization of the protective layer could repassivate.

An important related issue is the accuracy in measuring very low general corrosion rates. General corrosion rates on the order of nm/year are



7.1 Mechanism of the breakdown of the passive film induced by enrichment of sulphur at the metal–passive film interface (Marcus, 1995). Used with permission from Taylor and Francis.

difficult to measure accurately. The accuracy is important because the rates must be extrapolated to a very long time period to calculate the extent of general corrosion and assess when the package would fail.

Once the passive film becomes unstable without repassivation during disposal, either high general corrosion or localized corrosion such as crevice corrosion or pitting corrosion would occur. For localized corrosion to be initiated, if there is no existing (propagating) pitting or crevice corrosion, the corrosion potential needs to reach the breakdown potential for highly corrosion-resistant alloys (such as nickel-based alloys) (Ahn *et al*., 2008b, 2013; ASM International,1993). This condition is determined by the severity of the evolved groundwater chemistry. More conservatively, at the corrosion potential below the repassivation potential, even the existing (propagating) pitting or crevice corrosion would be arrested. The breakdown potential or repassivation potential generally increases with higher concentration ratios of oxyanions such as nitrates to chloride (Dunn *et al*., 2005). Even if the localized corrosion occurs, it is not expected to open up entire areas of a container surface. The cathodic capacity of the outside of an active crevice or pit, from the separated cathodic area from the active area, would limit localized corrosion propagation fronts (Shukla *et al*., 2007). Some studies show only pit growth rather than uniform dissolution in the crevice area of highly corrosion-resistant alloys (Ahn *et al*., 2008a). Based on the cathodic capacity limitation, a maximum of 20% of the surface area is likely to be open (He *et al*., 2011).

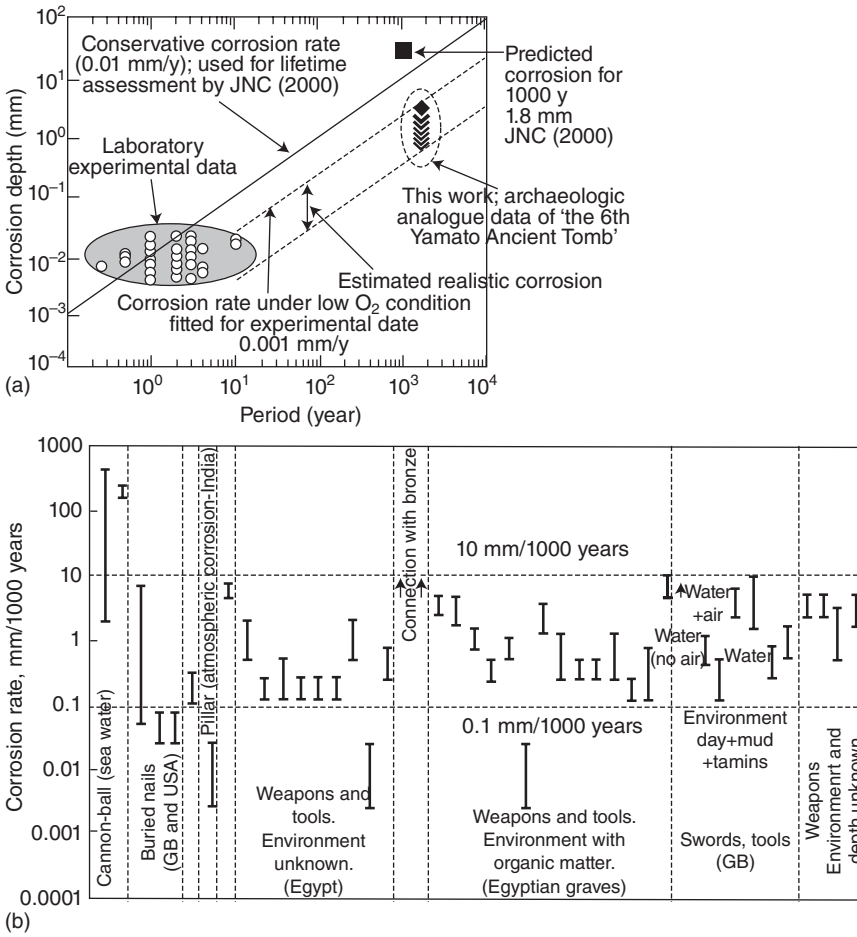
7.3.2 Carbon steel corrosion in mild reducing aqueous environments

Carbon steel is a corrosion-allowance metal that is expected to have a relatively low corrosion rate in a mild, near-neutral pH, and reducing environment such as granite and clay (Jung *et al*., 2011). One localized corrosion process in carbon steel is pitting corrosion. This pitting process is empirically represented by a 'pitting factor', which is defined as a ratio of pit penetration depth to the uniform corrosion depth. Therefore, degradation by pitting corrosion of a carbon steel container can be represented by adjusting the magnitude of the general corrosion rates.

Even in an underground repository, which is planned in the long term to have a reducing environment, it will initially have an oxidizing condition due to the excavation conducted before closure (which provides oxygen). The general corrosion rate of carbon steel in an oxidizing environment is very high: in the range of 10–100 μm/year (3.94×10^{-4} to 3.94×10^{-3} inch/year) at room temperature in simulated mild initial groundwater (Jung *et al*., 2011). During 30-year oxidizing conditions applicable for carbon steel corrosion, a general corrosion rate of 50 μm/year (1.97×10^{-3} inch/year) will

result in a small penetration depth of 0.15 cm (0.02 inch). Estimating the carbon-steel general corrosion rate using laboratory data and analogue data indicates corrosion rates in the range of 0.1–10 μm/year (3.94×10^{-6} to 3.94×10^{-4} inch/year) in the reducing environment (Yoshikawa *et al.*, 2008; David *et al.*, 2002). Figure 7.2 shows the data collected from Yoshikawa *et al.* (2008) from Japan and David *et al.* (2002) from France.

Carbon steel is susceptible to pitting corrosion in an oxidizing environment. In addition, carbon steel is expected to corrode at a higher general

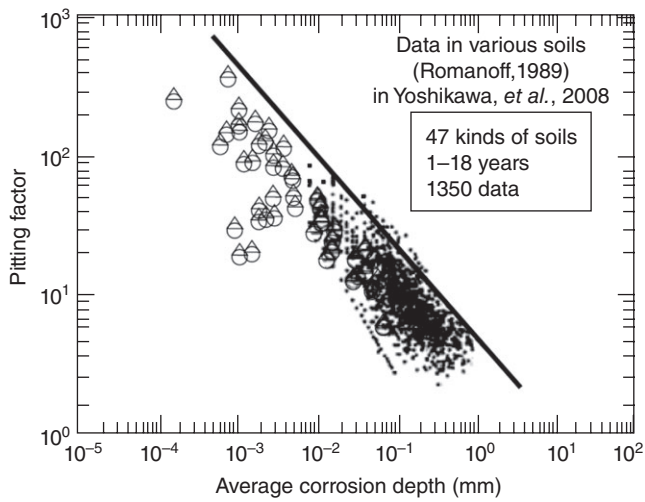


7.2 Measured corrosion rates of carbon steel in simulated solutions and correlation with archaeological analogue data up to 1,000 years: the first analysis was conducted in Japan (a) (used with permission from Elsevier), and the second in France (b) (used with permission from Maney Publishing).

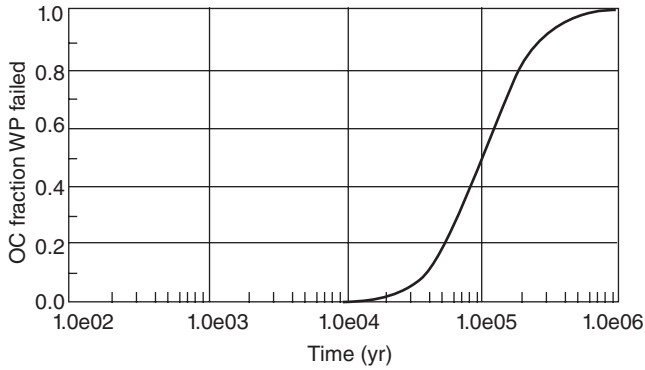
rate in an oxidizing compared to a reducing environment. However, the effect of pitting corrosion can be accounted for by using an enhanced general corrosion rate. The pitting factor, which is a ratio of pit propagation depth to general corrosion penetration depth, will approach unity during the oxidizing period as the general corrosion proceeds deeper. The pitting does not enhance the corrosion penetration at this point. An example case is shown in Fig. 7.3 (Johnson and King, 2000).

7.3.3 Risk insights of general corrosion and localized corrosion

Container failure by general corrosion is likely to result in sufficient opening of the container surface to allow substantial advective release of radionuclides. The rate of release of radionuclides by advective release is expected to be higher by several orders of magnitude than diffusive release that may occur through tight cracks or small pits. Therefore, underestimating the general corrosion rates because of the uncertainties may lead to an inaccurate, delayed and low-magnitude radionuclide release from a failed container. When the uncertainties associated with the general corrosion rates are random in nature, the general corrosion rates are expressed in a uniform distribution, either in a linear scale or in a log scale, depending on characteristics of the uncertainties. Figure 7.4 is an example output of the failure probability of a carbon steel container with time, using the range of general



7.3 Variation of the pitting factor for carbon steel with the average depth of corrosion derived from long-term corrosion tests and short-term laboratory measurements (Johnson and King, 2008). Used with permission from Elsevier.



7.4 Carbon steel waste package failure time for geologic disposal system in reducing environment (Jung *et al.,* 2011). Used with permission from American Nuclear Society (ANS).

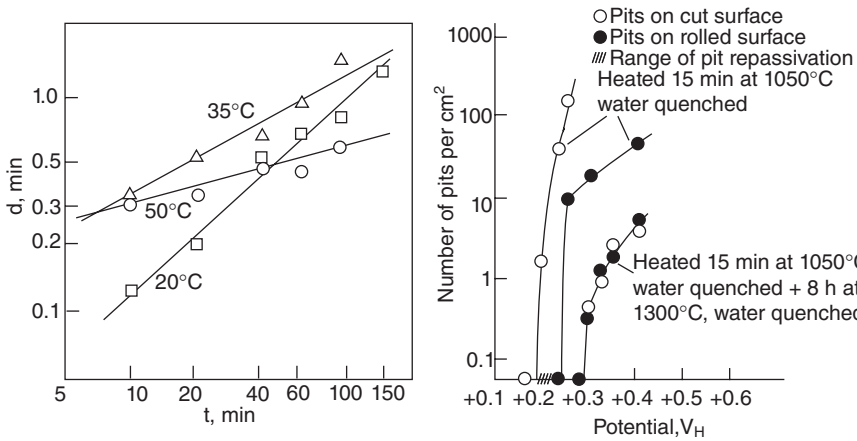
corrosion rates sampled from a log normal distribution extracted from Fig. 7.2 (David *et al*., 2002; Jung *et al*., 2011). The radionuclide release at a given time will begin from a finite number of containers that failed, from calculations using the probability of failure and the total number of containers.

Container failure by localized corrosion may also limit the radionuclide release because of restricted flow through the small perforations due to the pits. Pit diameters were from micrometers to millimeters (0.4 microinch to milliinch) and pit density is 0.1–100/cm^2 (0.6–645/inch^2) as shown in Fig. 7.5 from selected metals and aqueous environments (Szklarsksa-Smialowska, 1986). In addition, the pits are usually filled with corrosion products or solid precipitates from groundwater. Therefore, radionuclide release through the restricted area is likely to be diffusive, which is generally slower than advective release.

7.4 Stress corrosion and hydrogen-induced cracking of carbon steel and stainless steel

This section presents the SCC behaviour of carbon steel disposal containers and stainless steel storage canisters. Carbon steel is mostly susceptible to hydrogen-induced cracking due to residual weld (and heat affected zone) stress or seismic-induced impact stress. Hydrogen is generated during general corrosion or gamma radiolysis of groundwater in a reducing environment (Ahn and Soo, 1995).

In a marine (coastal) environment, salt deposits may occur on the stainless steel canister surface due to salt deposits in the humid air. The salt deposits on the canister when the canister surface temperature is above ambient. Aqueous conditions of high chloride concentration may form due to this salt deliquescence. With the residual tensile stress at welds (including

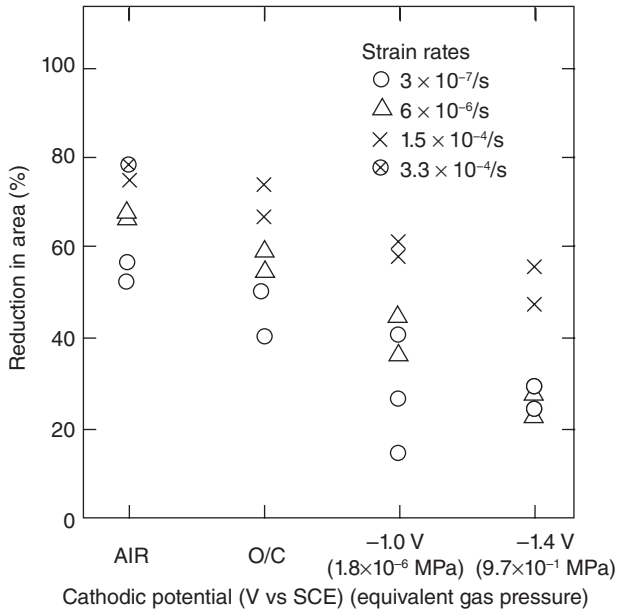


7.5 Examples of size and distribution of pits. Pit diameter vs time for 18Cr-12Ni-2Mo-Ti stainless steel in 0.1 n H_2SO_4 + 0.1 n NaCl. Normal pit size observed is in the range of micrometers to millimeters and pit density is for 304 stainless steel after potentiostatic polarization in 1 m NaCl solution. The term 'd' is pit diameter and 't' is time (reprinted with permission) (Szklarsksa-Smialowska, 1986). Used with permission from NACE International.

heat affected zone), this high concentration of chlorides may induce SCC (Shirai *et al.*, 2011; EPRI, 2005). SCC can be screened out based on stress mitigation techniques (or other remediation) such as applying compressive stress in the weld. In the absence of this mitigation, the opening surface area by SCC (or hydrogen-induced) cracks may be estimated, if a canister were susceptible to SCC. This quantitative estimate allows assessment of the radionuclide release due to waste form degradation inside the container or the canister.

7.4.1 Hydrogen-induced cracking of carbon steel

Carbon steel may be susceptible to hydrogen-induced cracking (Kobayashi *et al.*, 2011). Hydrogen is likely to be produced by radiolysis of groundwater and by water reduction during the corrosion process in reducing conditions. Figure 7.6 shows reduction in area versus applied potential in constant extension rate testing (CERT) for ASTM A216-Grade WCA steel in concentrated synthetic groundwater at 80°C (176°F) (Ahn and Soo, 1995). This hydrogen-induced cracking can be regarded as a variation of SCC. Recently, the surface opening area resulting from SCC container damage has been assessed for various candidate container metals including carbon steel. Generally, the maximum opening area is approximately 0.1% of the total surface area of the waste package (Gwo *et al.*, 2011). This original



7.6 Reduction in area versus applied potential in constant extension rate testing (CERT) for ASTM A216-Grade WCA steel in concentrated synthetic groundwater at 80°C (Ahn and Soo, 1995). SCE: standard calomel electrode. Used with permission from Elsevier.

assessment was made with impact stress in the deformed area of the container as an effect of seismic impact. However, this assessment may be applicable to the normal static case too. The weld residual stress and a weld area can be used instead of impact stress and deformed area from seismicity. The models for estimating opening area due to SCC are described in the following section for stainless steel canisters. This approach is equally applicable to the hydrogen-induced cracking of carbon steel.

7.4.2 SCC of stainless steel

Salt deposits on the canister surface open to the environment may be significant in coastal areas. SCC of the stainless steel canister needs to be considered when the relative humidity (RH) in air is appropriately high, the amount of salt deposits is sufficient to form aggressive and sufficient aqueous conditions at welds, and when a sufficient tensile stress is present. If the RH is too low, the aqueous condition would not exist. On the other hand, if RH is too high, the chloride concentration would not be high enough to initiate SCC. The weld area could have residual tensile stress and sensitized microstructure which is prone to SCC.

The RH of the environment surrounding the canister surface and salt deposits depends on the canister surface temperature. Over a long time, the surface temperature will decrease as the radioactivity inside the canister gradually decays. This will result in increasing RH of the environment immediately adjacent to the canister surface. Also, temperature, RH and the amount of salt deposits will not be homogeneous on the canister surface because of the SNF storage configuration and air flow surrounding the canister. In addition, primarily the weld areas will be susceptible to SCC. Considering these environmental and materials factors, the probability associated with SCC could be low enough for it to be screened out from performance assessment (PA), especially with appropriate remediation.

If SCC were to occur, radionuclide releases may be primarily caused by the release of aerosol radioactive materials, which may in turn be driven by the pressure of inert fill gas and fission gas inside the canister (from prior release from failed cladding). The release rates are also affected by the opening area of the canister surface caused by SCC. The SCC area density per weld area of the canister may be estimated conservatively (the estimate was originally under seismic events) by the following equation (Gwo *et al*., 2011):

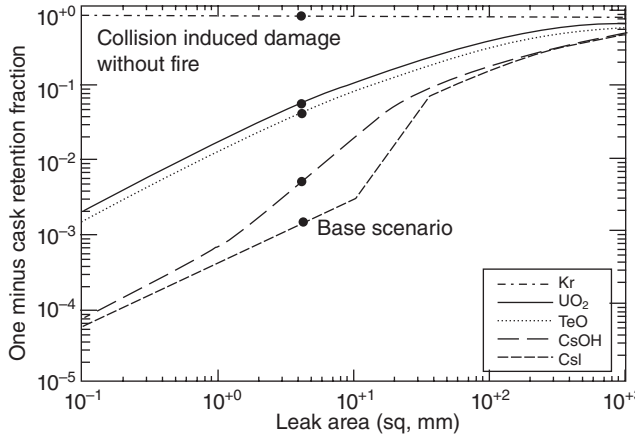
$$\delta = C\,\sigma / E \qquad [7.1]$$

where δ is crack areal density (m^2/m^2), σ is applied stress (MPa), E is Young's modulus (MPa) and C is geometric constant.

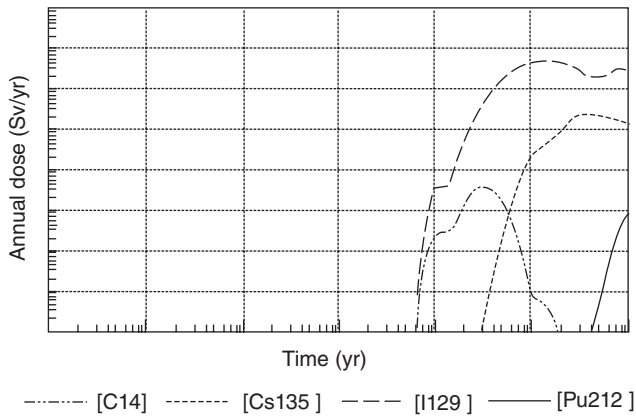
For example assuming no inspections and remediation, a calculation for stainless steel using Eq. [7.1] suggests that the crack areal density per unit weld area is approximately 1.2×10^{-3} at 170–310 MPa (25–45 ksi) of applied stress, (193–207) $\times$ 10^3 MPa [(28–30) $\times$ 10^3 ksi] of Young's modulus (Gwo *et al*., 2011). The weld area fraction is about 10^{-2}–10^{-1} of total surface area (ASM International, 1993). In a canister surface area of about 30 m^2 (4.6×10^4 $inch^2$), the surface opening area will become $3.6 \times (10^2$–$10^3)$ mm^2 (0.56–5.6 $inch^2$). The model in Eq. [7.1] is conservative, assuming a distribution of uniform crack size. In reality, the number and size of cracks are likely to be smaller. This calculated area is obviously larger than that allowed for leak tightness (Institute for Nuclear Materials Management, 1997).

7.4.3 Risk insights of the cracking of carbon steel and stainless steel

Figure 7.7 shows an example of the radionuclide release fraction to the environment from a cask (Sprung *et al*., 2000). Casks include canister and other overpacks. Strictly speaking, this figure was constructed for a transportation cask. Nevertheless, the radionuclide release behaviour would be similar in the storage cask. The radionuclide release fraction is expressed by (1.0 – Retention). In this range of the surface opening area, the surface



7.7 Cask-to-environment release fractions (1.0 – Retention) versus open cask surface area (Sprung *et al.*, 2000).



7.8 Example β-SOAR (Markley *et al.*, 2011) dose results for only commercial SNF using combined degradation rate in a stylized reducing geological disposal system (Ahn *et al.*, 2011a). Used with permission from American Nuclear Society (ANS).

opening is already wide enough to result in the release fraction approaching to 1. This retention mechanism is in addition to the low radionuclide release fraction from the degraded UO_2 matrix and the failed cladding. In reality, the surface area opening by SCC may be smaller because the model of Eq. [7.1] is conservative.

Figure 7.8 (calculated using the β-SOAR model of Markley *et al.*, 2011) shows an example of a calculation of radionuclide release from the seismic-induced SCC of various disposal containers (Gwo *et al.*, 2011). There is an additional factor lowering the magnitude of radionuclide release due to the restricted perforation made by SCC.

7.5 Spent nuclear fuel (SNF) degradation

This section presents the degradation behaviour of SNF in mild and near-neutral environments under (i) oxidizing or reducing aqueous disposal conditions, and (ii) in dry storage environments. During the aqueous dissolution of SNF, highly soluble fission products such as Tc-99 or I-129 are released congruently with (i.e., in proportion to) the SNF matrix (UO_2) dissolution. On the other hand, actinides such Pu-239 or Np-237 are released at a concentration below or equal to their solubility limits (or colloid concentration), which are in turn determined by the SNF matrix dissolution rate, groundwater flow rate and solubility limit. Colloids are suspended solid particles of less than 1 micrometer in size that can contain actinides. An oxidizing aqueous environment promotes electrochemical dissolution of the SNF matrix in soluble species with the aid of oxidants such as dissolved oxygen and hydrogen peroxide (Shoesmith, 2000). In a reducing environment, the UO_2 matrix will dissolve chemically in soluble species (Sunder and Shoesmith, 1991). Generally, the electrochemical dissolution rate is faster than the chemical dissolution rate. In the presence of radiolysis effects, the SNF matrix may dissolve in either an electrochemical or a chemical process, depending on the magnitude of the radiolysis (Ahn *et al*., 2011a). In conjunction with container failure and sorption and/or flow behaviour of backfill, the SNF matrix dissolution serves as the source term of radionuclide release in the PA. In a dry storage environment, mechanical degradation of the SNF matrix could occur by air oxidation/humid air hydration or impact fragmentation upon the canister failure under normal conditions (e.g., SCC failure) or external hazard conditions (e.g., aircraft or seismic impact). In the canister, if incomplete drying of SNF assemblies occurs, the residual water may increase RH sufficiently to oxidize (by oxygen from the radiolysis of water molecules) or hydrate the SNF matrix. With severe external hazards, high temperatures or impact stress may fragment the SNF matrix by oxidation or mechanical disintegration. The respirable SNF particles (i.e., suspended aerosol, less than 10 μm [3.9 microinch] in size) produced by the fragmentation serve as the primary source term for radionuclide release in air.

7.5.1 SNF dissolution

The dissolution rates by the electrochemical and chemical processes are (Ahn, 1996a):

$$R_{dis} = \frac{S}{V} k_e f(E) \qquad [7.2]$$

and

$$R_{dis} = \frac{S}{V} k_{-}(C_s - C_t) \quad [7.3]$$

$$R_{dis} = \frac{A}{V} k_{+}(C_t - C_0) + \frac{F}{V} C_t + N_{par} C_t \quad [7.4]$$

where S is surface area of the dissolving phase, V is leachate volume, k_e is rate constant for electrochemical dissolution, $f(E)$ is dissolution rate as a function of electrochemical potential E, k_{-} is rate constant for SF dissolution, C_s is effective solubility limit of dissolving phase, C_t is elemental concentration under consideration, k_{+} is rate constant for growth of the reprecipitated phase, F is flow rate of ground water, and N_{par} is formation or growth rate of colloids per unit leachate concentration.

Equation [7.2] is for electrochemical process, Eq. [7.3] is for chemical process, and Eq. [7.4] is for release rate from the dissolution processes of the first two equations.

The fractional mobilization rate is the dissolution rate multiplied by the specific surface area of the SNF matrix. Conservatively, the fractional mobilization rates can be assumed constant within uncertainty ranges at a given temperature. The environmental conditions are important in determining the dissolution rates, including near field water chemistry, temperature, pH, or reducing or oxidizing conditions. Important water chemistry includes carbonates, and cations such as calcium or silica species (Ahn and Mohanty, 2008).

In connecting the dissolution rate to the fractional mobilization rate, the specific surface area is determined by the average fragment size (radius) and density of the waste form. Typically, the fragment size of commercial SNF is 0.1 cm (0.04 inch) (Ahn and Mohanty, 2008).

If the temperature exceeds 100°C (212°F), solid-state oxidation or hydration will occur, depending on the RH. Higher uranium oxides ($UO_{2.4}$ or U_3O_8) that form by oxidization of the UO_2 matrix dissolve at a rate similar to the unoxidized UO_2 matrix. Hydrated $UO_3{\cdot}xH_2O$ (x = 0.8, 2) dissolves 10–20 times faster than unhydrated oxides. However, the rate of hydration (i.e., the formation rate of $UO_3{\cdot}xH_2O$) is slower than the aqueous dissolution rate. Ahn and Mohanty (2008) summarized the effects of oxidation and hydration on the dissolution.

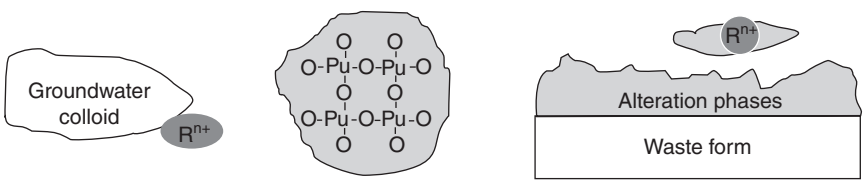
7.5.2 Colloid formation and solubility limit

Actinides such Pu-239 or Np-237 have low solubility limits, and they are released at a concentration below or equal to their solubility limits (or colloid concentrations), which in turn are determined by the SNF matrix dissolution rate, groundwater flow rate and solubility limit.

Regarding colloid formation, Ahn (1996a) summarized the processes involved. During the dissolution of the SNF matrix, suspended solid particles containing mainly actinides of low solubility may form. The colloids can carry a large amount of actinides compared with dissolved species. The traditional processes of colloid formation (especially in actinide colloids) have been investigated under near-equilibrium conditions. Most studies in this regard pertain to chemical bonding among ions. Extending the chemical bonding process in equilibrium or non-equilibrium states, colloid formation may be described in macroscopic ways by three different processes: (a) condensation, (b) dispersion, and (c) sorption (pseudo-colloid formation). Colloids may form by precipitation in small particles because of supersaturation of actinides or the SNF matrix (i.e., condensation). The layer of the precipitated phases on the SNF matrix can be mechanically detached into small suspended particles (i.e., dispersion). Finally, the dissolved pure actinide species can be sorbed on the surface of non-radioactive inert groundwater colloids (i.e., sorption). Figure 7.9 shows schematics of these three processes (CRWMS M&O, 2001).

7.5.3 Dry oxidation or hydration, and mechanical fracture

Dry-air oxidation or humid air hydration of SNF in air or in the presence of limited amounts of groundwater may play an important role in radionuclide releases (Ahn and Mohanty, 2008; Ahn, 1996b). The UO_2 matrix will fracture (or crack) upon oxidation or hydration by volume change. Lower oxidized oxides such as $UO_{2.4}$ will contract, whereas higher oxides such as U_3O_8 or UO_3 hydrates will expand. Lower and higher oxides are defined here as oxides with a (O/U) ratio smaller and larger than 2.4, respectively. Fractions (e.g., 10^{-6}–10^{-3}) of oxidized or hydrated phases are likely to be respirable aerosol less than 10 micrometer (3.9 microinch) in size. The aerosol will increase the radionuclide release in air. The oxidized or hydrolysed phases also increase the area of SNF surface exposed to groundwater. This increase of the exposed surface area is in turn expected to increase radionuclide release in groundwater. Similarly, mechanical impacts such as those caused by seismic events can also fragment the SNF into particles.



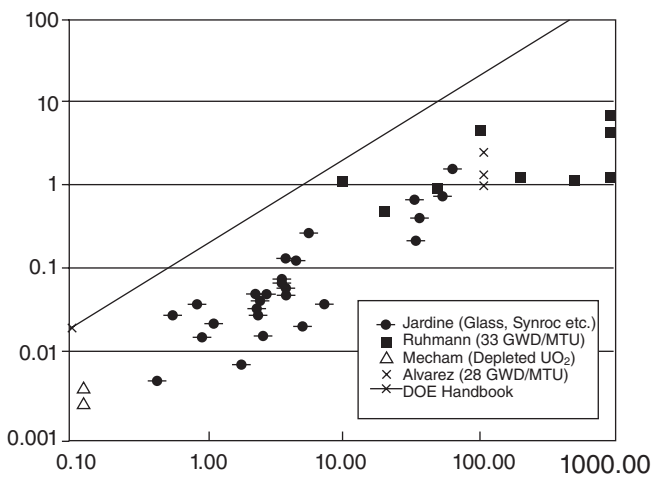
7.9 Types of colloid formation (CRWMS M&O, 2001).

Figure 7.10 shows the fraction of respirable particles, depending on the impact energy absorbed. The fine-grained and porous rim structure near cladding of high burnup (above about 60 GWd/MTU) UO_2 may also affect the magnitude of the radionuclide release fraction (NRC, 2007).

7.5.4 Risk insight of SNF degradation

Table 7.1 summarizes the dissolution rates for oxidizing and reducing disposal environments (Ahn *et al.*, 2011a) used in a performance assessment model (Markley *et al.*, 2011). A range of environmental conditions are considered, mostly near-neutral pH and ambient temperature. The variation of pH and temperature can be adjusted in terms of dissolution rate as user-defined parameters. For this base case, radionuclide release is estimated combining the reducing and oxidizing environments, to simulated residual radiolysis of water by actinides in the reducing environment. Figure 7.8 shows the estimated dose from the radionuclide release for this combined case.

Considering all radionuclide release fractions from the UO_2 matrix, an exercise was conducted to estimate the doses to workers or members of the public from airborne fragments of the SNF matrix caused by SNF oxidation and SNF drop/collision (after Kamas *et al.*, 2006). The most significant dose contributor in the release fraction is aerosol SNF fines (i.e., small solid particles). Tritium, noble gases, iodine, crud, ruthenium, caesium, strontium and SNF fines were part of the source term considered. In Fig. 7.11, the

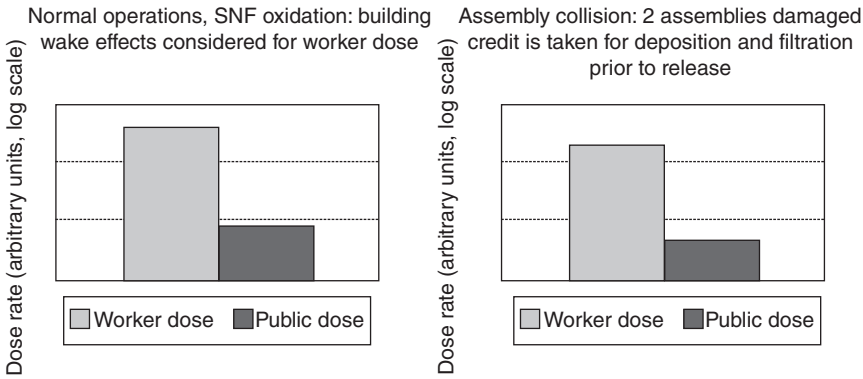


7.10 Comparison of the DOE handbook respirable fraction equation to experimental values of the specific energy input into the brittle material (NRC, 2007).

Table 7.1 Summary of SNF dissolution rates in oxidizing and reducing environments

Parameter name	Value	Description and basis
Mobilization (degradation) of commercial SNF and sMOX (spent MOX) under the oxidizing condition (fraction per year; minimum and maximum)	3.00E-05, 6.00E-04 (log-uniform)	The oxidizing environment is considered because of the potential alpha radiolysis in the reducing environment and the early waste package failure. The assessment is more based on immersion conditions that are considered in the alternative disposal sites in the future.[9] The dissolution rate of commercial SNF is assumed to be bound to that of sMOX under immersion conditions.[12] Both commercial SNF and sMOX have the particle size of ~1 mm after reactor irradiation. Other references include the references of [3] and [7].
Mobilization (degradation) of commercial SNF and sMOX under the reducing condition (fraction per year; minimum and maximum)	9.00E-07, 2.00E-05 (log-uniform)	An average factor of 0.03 (0.01–0.1) was factored in the oxidizing case. In the French and Belgian repositories, an average 2×10^{-6}/year was used[13], similar to the current estimate. To be consistent, the dissolution rate of sMOX was assumed to be the same as the rate of commercial SNF[12].
Mobilization (degradation) of commercial SNF and sMOX under the combined condition (fraction per year; minimum and maximum)	9.00E-07, 6.00E-04 (log-uniform)	Because the alpha radiolysis may have limited effects on the dissolution rate of commercial SNF[14] and sMOX, the combined case is separated to represent some effects of alpha radiolysis. If we consider the hydrogen effects to be produced by the container corrosion, this combined rate could be conservative. The hydrogen could inhibit the SNF dissolution rate.[15] To be consistent, the dissolution rate of sMOX was assumed to be the same as the rate of commercial SNF.[12]

Spent MOX fuel is also included in the table and the reference numbers quoted are from the reference by Ahn *et al.* (2011a).



7.11 Example dose estimate for (a) oxidation and (b) collision (/drop) of SNF assemblies (after Kamas *et al.,* 2006). Used with permission from American Nuclear Society (ANS).

radionuclide release fraction of the aerosol SNF fines, 2.0×10^{-6} for the drop/collision case and 1.2×10^{-3} for the SNF oxidation case, were used to estimate the dose to workers or members of the public (Ahn *et al*., 2011b; Kamas *et al*., 2006). A site boundary was defined, for the dose to workers within the boundary and to members of the public outside the boundary.

The left figure is for SNF oxidation under normal operations. The wake effects are a modification of the radionuclide transport path right outside any storage building if any building shadow exists. Consequently, radionuclide transport will stop. Within a short distance from the building, the radionuclide transport will not be reached. The right figure is for drop/collision cases. In both cases, arbitrary dose rate units are used for the log scale. The oxidation case gives a dose rate ten times higher than the collision case in the same log-scale unit.

7.6 Cladding performance

This section presents the performance of cladding in aqueous disposal environments and dry storage environments. Hydrogen-induced cracking of cladding may be a major detrimental degradation mechanism for both disposal and storage conditions. Crack opening area allows radionuclide release under both conditions. Oxidation (or general corrosion) of cladding is very slow and localized corrosion is unlikely to occur in near-neutral pH disposal environments (Ahn, 1996b). Oxidation of cladding is only possible in the presence of residual water and/or oxygen in dry storage canisters. Initially defective cladding may be further cracked (unzipped) by the pressure imposed on it by corrosion products of the SNF matrix or zirconium

itself. Longer longitudinal cracks that develop from the initial cracking/unzipping will increase radionuclide release under both conditions.

7.6.1 Hydrogen effects

During reactor operations, the cladding metals, mainly Zircaloy, corrode in water. This introduces hydrogen into the Zircaloy. Hydrogen can degrade the strength of Zircaloy through overall embrittlement caused by a dispersion of radially oriented hydrides (perpendicular to the hoop stress) (Chung, 2004). The hydrides formed during reactor operations are mostly circumferential hydrides (parallel to the hoop stress). Circumferential hydrides may not affect the strength significantly, depending on the magnitude of severity. However, circumferential hydrides are known to become radially reoriented in the presence of appropriate applied stress and temperature (Chung, 2004). Figure 7.12 compares hydrides oriented circumferential or perpendicular to the hoop stress and Fig. 7.13 shows ductility loss with radial hydrides (Yagnik *et al.*, 2004).

Another hydrogen effect is delayed-hydride cracking (DHC). Small cracks that develop on the inner or outer surface of cladding may lead to crack propagation when assisted by hydrogen diffusion to the crack tip, thus forming radially oriented hydrides at the crack tip. The mechanism has not been proven to exist under dry storage conditions. Figure 7.14 shows a schematic for the mechanism of the DHC process. The crack density and size from hydrogen embrittlement of hydride reorientation and DHC can be conservatively assessed like the SCC of stainless steel described in Section 7.4.2.

7.6.2 Unzipping of cladding

A maximum of 1% of SNF discharged from reactors could be defective. Volume expansion associated with the oxidation/hydration of the SNF matrix or zirconium may crack/unzip defective cladding (Cunnane *et al.*, 2003). Figure 7.15 shows a schematic of this unzipping process (DOE, 2002). Unzipping was observed in the Argonne National Laboratory 1.5-year long tests, caused by stress generated by corrosion product accumulation in the gap of cladding and the fuel matrix from uniform corrosion of Zircaloy cladding at 175°C (347°F) (Cunnane *et al.*, 2003).

Oxidation/hydration may occur with either residual moisture inside the intact canister or container, or from moisture that has intruded into the failed canister or container. This cladding failure may affect the magnitude of the radionuclide release fraction and challenge the retrievability of the SNF materials, and lead to configuration changes in internal structure that impact nuclear criticality.

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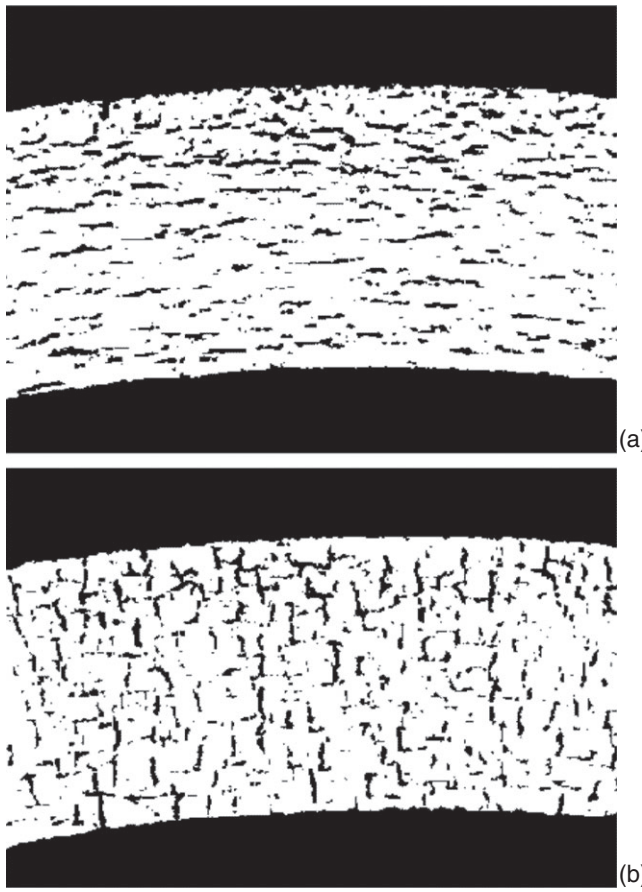
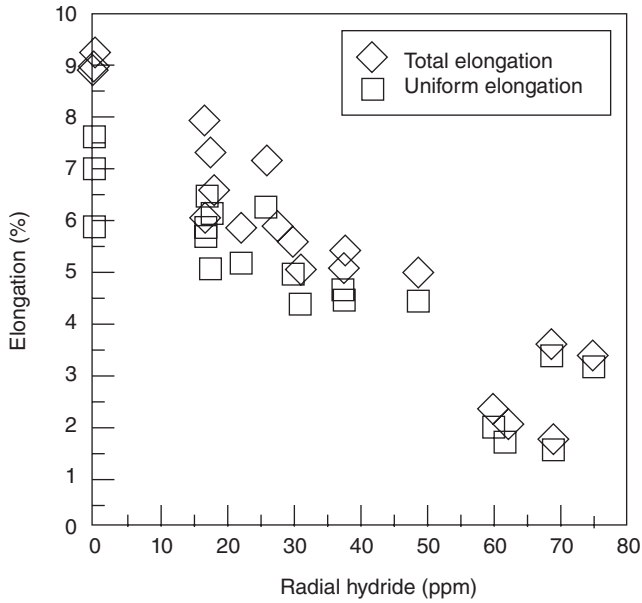


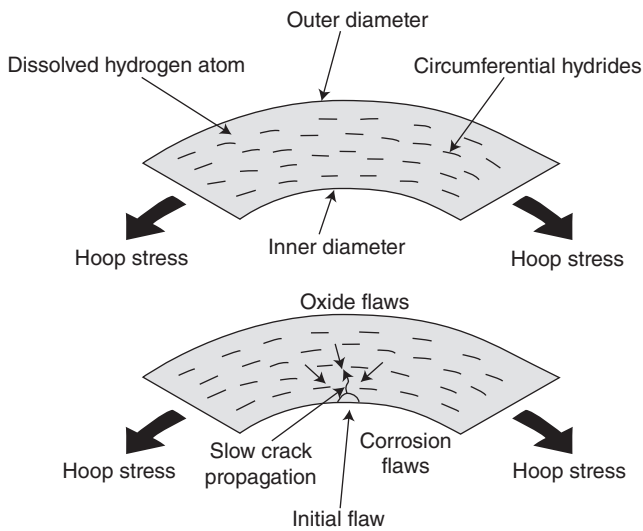
7.12 Hydride reorientation from circumferential (a) to radial (b) direction to hoop stress (Yagnik *et al.*, 2004); cladding thickness of ~0.6 mm. Used with permission from American Nuclear Society (ANS).

7.6.3 Risk insight of cladding performance

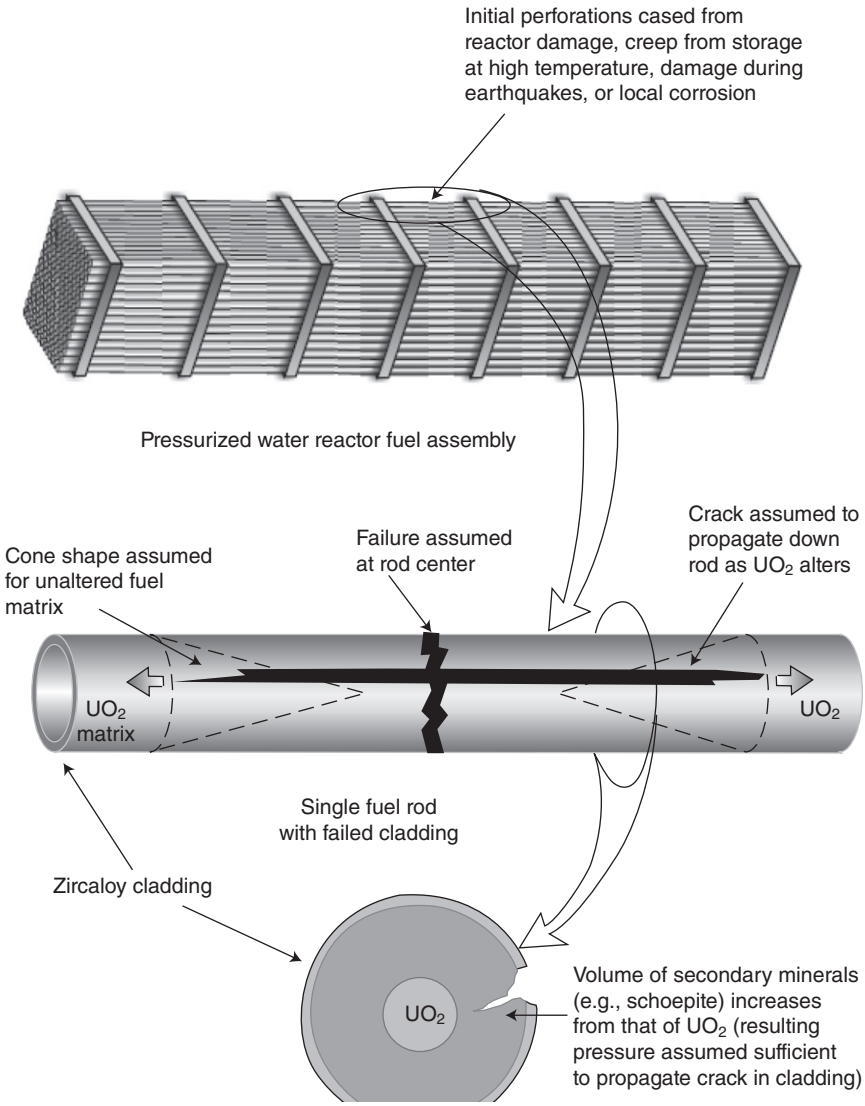
The values of crack opening from models need to be compared with those used in determining the radionuclide release fraction in storage from experimental work (Lorenz *et al.*, 1980). Lorenz *et al.* (1980) conducted burst tests by heating a clad SNF rod, allowing an opening area of about 1.6 cm^2 ($\sim 10^{-1}$ fraction of the total cladding surface). If the calculated value of the cladding area opened by cracking resulting from hydrogen effects is smaller than that from the experiments by Lorenz *et al.* (1980), the radionuclide release fraction will not increase with further cladding cracking from the embrittlement. Otherwise, the radionuclide release fraction from the UO_2 matrix to the canister inside will be affected by the embrittlement. The current



7.13 Effects of radial hydrides on ductility (elongation) loss in a more severe case. The different orientations result in different magnitudes of ductility loss (Yagnik *et al.*, 2004). Used with permission from American Nuclear Society (ANS).



7.14 A schematic of the DHC process showing hydrogen diffusion to an existing crack tip. The threshold stress intensity factor for DHC of Zircaloy is in the range of 5 MPa $m^{1/2}$. Under storage conditions, such level of stress intensification has not been demonstrated, although a few possibilities are currently under study.



7.15 Cladding unzipping process by the oxidation/hydration of the SNF matrix or zirconium (DOE, 2002).

regulation for SNF storage requires that the cladding must be protected during storage against degradation that leads to gross rupture or the SNF needs to be otherwise confined.

The effects of crack opening on the radionuclide release in disposal were also studied. Ahn and Mohanty (2008) summarized the literature results. In the presence of partial protection from failed cladding by cracking, the

dissolution rate decreases significantly compared to bare SNF, with slit (~0.015 cm [5.9 × 10 inch] width and ~2.54 cm [1 inch] length) or hole (~0.02 cm [7.9×10^{-3} inch] diameter) defective SNF cladding under immersion conditions in J-13 well water at 85°C (185°F). The tests were intended to simulate cladding partially failed by localized corrosion or SCC. The radionuclide release rates decreased by a factor of ~140 for Tc-99, ~7×10^5 for I-129, and ~65 for Sr-90, compared to bare clad SNF. However, in the segment tests of clad SNF under immersion conditions, the radionuclide release rates did not decrease compared to bare SNF. In these tests, nearly half of the surface area was exposed. Similar conclusions can be drawn from tests with Canadian deuterium-natural uranium reactor clad SNF, immersed in Canadian granitic groundwater. These results suggest that cladding would not inhibit the dissolution rates if it fails catastrophically exposing a substantial surface area to the solution (groundwater).

Elam *et al*. (2003) assessed the effects on nuclear subcriticality caused by the configuration changes due to cladding failure. The main assumption in this study is full water flooding in the cask. The report presented the reactivity for various SNF rod conditions. For uniform burnup of 45 and 75 GWd/MTU collapsed SNF rods, the variations of the neutron multiplication factor, Δk_{eff}, were not significant for this changed cladding configuration.

7.7 Summary

This chapter presents examples of analytic approaches and methodologies for modelling the behaviours of waste forms and waste package metals in long-term management of spent nuclear fuel (SNF) and high level waste (HLW). Two cases, long-term geological disposal, and interim extended dry storage, were considered.

Selected important topics in the management were presented on:

- long-term integrity of passive film, slow general corrosion, and localized corrosion in disposal container: the role of passive film was discussed in terms of long-term slow general corrosion, and fast localized corrosion upon the passivity breakdown in corrosion resistance of metals, and slow general corrosion of corrosion allowance of metals in the reducing environment.
- SCC/hydrogen embrittlement of carbon steel and stainless steel was discussed with respect to their performance in disposal or storage.
- Spent nuclear fuel degradation was discussed in terms of radionuclide release in disposal and storage systems.
- Cladding performance was discussed in terms of radionuclide release and criticality control in disposal and storage systems.

In each topic, the involved risk insights were also discussed in the system performance assessment in disposal and storage.

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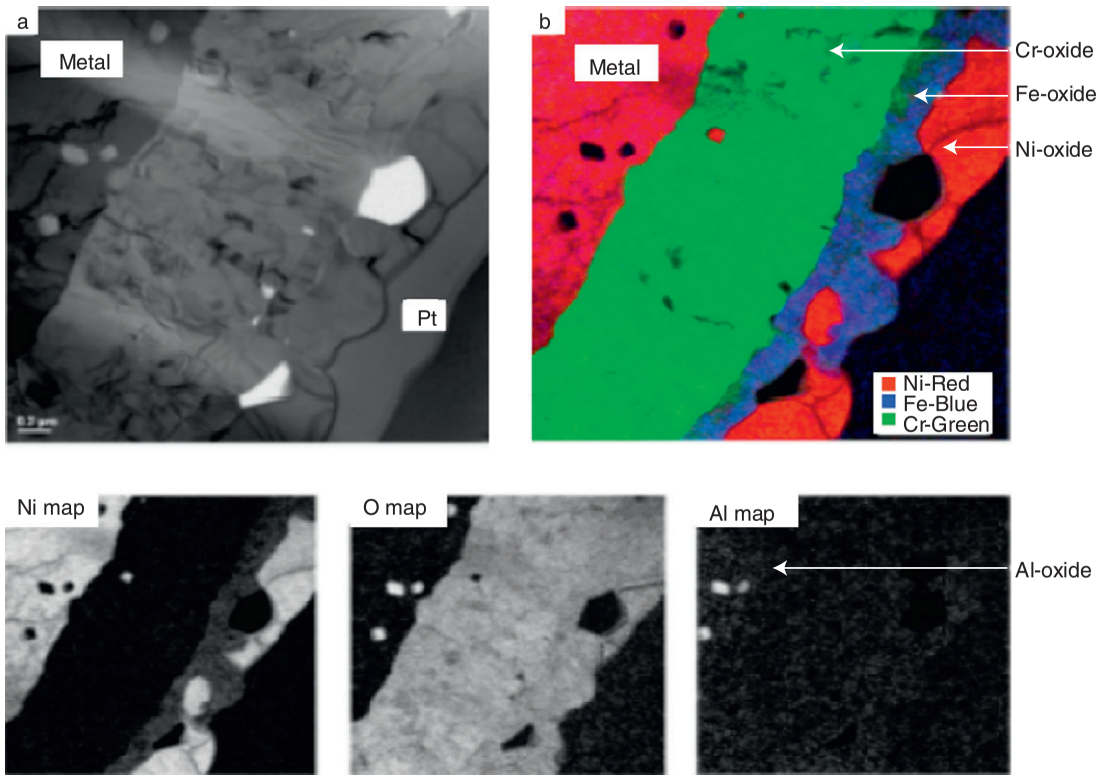

Plate II (Chapter 7) A cross-sectional view of passive film form on the surface of annealed nickel-based alloy (Alloy 22, Ni-22Cr-13Mo-4Fe-3W) (Orme, 2005).

8

Remediation of radioactively contaminated sites and management of the resulting waste

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Abstract: A large number of areas that have been contaminated by residual radioactive material as a result of past activities or accidents require remediation. These areas may be very large and of specific interest and may call for major commitments of funding and personnel resources. Environmental contamination may represent a hazard to the general public and the environment. This chapter provides information on protective and remedial actions that are intended to reduce existing or potential exposures and other impacts. Actions include the removal of the source of exposure, or other means such as restrictions of land use. Many factors should be considered during planning and implementation of environmental remediation, and are dealt with in this chapter, including site characterization, regulatory responsibilities, end state considerations, an appropriate selection of technologies, waste generation, management and disposal, and organizational measures. Current developments and trends are highlighted.

Key words: accident, clean-up, decommissioning, disposal, environmental remediation, planning, transportation.

8.1 Introduction: definition and extent of the problem

The situations dealt with in this chapter are interventions for areas that have been contaminated as a result of human activities and that could cause prolonged radiation exposure. In this context, the term 'areas' is used in its broadest sense and can include land, forests, urban environments and industrial sites. These areas may have been contaminated as a result of inadequate practices for radioactive waste management and disposal, radioactive discharges to the environment that did not meet regulatory requirements, nuclear accidents, atomic weapon tests, incidental releases of radionuclides by users of radioactive material or past practices that were not adequately controlled. This chapter also applies to radioactive discharges from facilities that were managed in accordance with less stringent requirements than those that are applied today (IAEA, 2006a). However, this chapter is not specifically intended for the management of huge amounts of uranium/

thorium mill tailings or naturally occurring radioactive material (NORM), which have their own specific circumstances and management options.

Some examples of contamination that might be encountered are given below. The list is not exhaustive but is intended to show the wide range of problems that might be found. Finally, although this chapter is intended for radioactively contaminated sites, the need for environmental remediation also includes non-radioactive, toxic contaminants that may be associated.

8.1.1 Nuclear power production and nuclear fuel cycle activities

The various stages of the nuclear fuel cycle and the operation and decommissioning of nuclear reactors all have the potential to create contaminated sites. The contamination may include spillage of ore end-product at the mine and in transport; waste from enrichment and fuel fabrication operations; fission product and actinide waste streams from reprocessing of fuel elements; radioactive effluents from normal operations of nuclear power plants; wastes produced during decommissioning of reactors; and major releases under accident conditions.

8.1.2 Production and use of radioactive substances for medical, research or industrial purposes

Radioactive materials have been used widely since their discovery for a variety of scientific, medical and industrial uses. In some cases, either through ignorance, carelessness or accident, sites have been left contaminated with residues of the operations. Such sites include, for instance, factories where radium was used in luminescent paint (see Chapter 15) and thorium was used in thorium-coated gas mantles.

8.1.3 Military activities and the production, testing and use of nuclear weapons

The manufacture of nuclear weapons involves the handling, transport and storage of large quantities of radioactive materials. The testing of weapons may involve nuclear yield and the release of fission products and activation products, or may involve the deliberate dispersal of radioactive materials in the environment. Some military use is made of depleted uranium which may contain fission products if obtained from reprocessed fuel. All of these activities have, in the past, resulted in contaminated sites, many of very large areas. Detailed examples are given in Part III of this book (Chapters 25–27).

8.1 Abandoned apartment building in the contaminated area near Chernobyl.

8.1.4 Major incidents

In the course of nuclear weapons production and transport, there have been several severe accidents resulting in considerable contamination. These include Windscale Pile 1 (1957); Kyshtym (1957); Palomares (1966); and Thule (1968). The spread of contamination from civilian industries by accident or by human ignorance are illustrated by the cases of Three Mile Island (1979), Chernobyl (1986) (Fig. 8.1), Goiania (1987) and Fukushima (2011). Table 8.1 shows a list of serious nuclear accidents rated according to the International Nuclear Event Scale (INES). It can be stated that accidents from level 5 up fall into the scope of this chapter (level 5 events necessitate at least some environmental control, and higher class events generally require environmental remediation). A comprehensive description of all the above-mentioned categories is given in Section 1.4.6 and IAEA (n.d.).

8.2 Planning and management of environmental remediation (ER)

To ensure that protective measures can be quickly and efficiently implemented to mitigate the adverse effects of an accident or other forms of long-term contamination at a nuclear site requires good planning, clear strategies and a good managerial team. Preparations for environmental remediation (ER) should, if possible, be done in two phases: preliminary planning, which should be available as part of normal operation or emergency preparedness for each nuclear facility; and detailed remediation planning, which takes into account site (and accident where applicable) specific

Table 8.1 Selection of nuclear accidents

Event site	Year	International Nuclear Event Scale (INES) rating	Impact on environment	Environmental remediation
Fukushima, Japan	2011	7	Extensive contamination, evacuation	Started
Chernobyl, Ukraine	1986	7	Extensive contamination, evacuation	Underway
Kyshtym, Russia	1957	6	At least 22 villages were exposed to radiation, a total population of around 10,000 were evacuated	Fragmentary information, contaminated soil was excavated and stockpiled in fenced enclosures
Goiania, Brazil	1987	5	Houses and scrapyards contaminated	Topsoil had to be removed from several sites, and several houses were demolished
Three Mile Island, USA	1979	5	Not lasting	Environmental control
Windscale, UK	1957	5	Not lasting	Environmental control

Level 7: Major accident. Major release of radioactive material with widespread health and environmental effects requiring implementation of planned and extended countermeasures.
Level 6: Serious accident. Significant release of radioactive material likely to require implementation of planned countermeasures.
Level 5: Accident with wider consequences. Limited release of radioactive material likely to require implementation of some planned countermeasures.

information. The two types of planning can be complementary and both are important in minimizing the detriment to society.

Life cycle assessment (LCA) is a systematic method used extensively for evaluating environmental effects of a technology or production process from cradle to grave. The method is gaining widespread acceptance in the field of supporting systems for environmental decision making. Indeed, the way environmental problems are seen and tackled by such an approach comes within the framework of sustainable development thinking. LCA actually enables listing and quantification of environmental burdens (radiological and non-radiological) and related impacts over the whole life cycle of a product, process or activity, from the very beginning to the end.

It is clear that LCA, when used by those who fully understand the technology being analysed, can be a useful addition to the process designer's toolkit. It ensures that pertinent questions are asked at the design stage and it can show where the environmental emissions/impacts occur and from where they arise within the process. If not addressed in the design stage, LCA can also be valid during on-going operations. With this knowledge, the process designer, or the operator of an existing process, can then set about minimizing these emissions/impacts, taking into account the overall life cycle of the operation. It is expected that early consideration and management of expected or on-going environmental impacts will reduce time and costs of remediation in comparison to actions taken only at the end of service life of a nuclear facility. A website devoted to LCA can be found in US EPA (2012).

From the radiation protection perspective, remediation projects are driven by two leading principles: justification and optimization. The first principle requires prior consideration of the benefit that would be achieved by the remedial action and also consideration of the harm it may eventually cause, in its broadest sense. In a very simple way, it can be stated that remediation should produce more good than harm. The second principle applies to the range of justified remedial options for which the net benefit would be positive. The optimum remedial option would be the one for which the net benefit is maximized. However, cost–benefit analysis methodology is limited to quantitative comparisons between the protection costs and the detriment costs. Some other approaches, namely multi-attribute utility analysis (see Section 4.5.1) allows the use of utility functions and introduces factors which are not easy to quantify in monetary terms as required in cost–benefit analysis (Fernandes *et al.*, 2013).

In many cases, the above principles are not fully considered in the decision-making process. Appropriate balance between the benefits and costs of remediation projects are particularly important in countries in which some sort of remediation programme is needed, as the costs of remediation (capital and labour costs) will have to compete with the benefits that the

application of that same amount of money would produce if directed to other purposes; for example, building schools or hospitals, or improving the existing infrastructure. It is clear that optimization may dictate the extent and end state of the remediation project, and so affect the generation and management of radioactive and other waste.

In addition to this, from the institutional perspective, it has to be kept in mind that remediation projects will entail longstanding administrative, monitoring and enforcement requirements; therefore the presence of solid institutions that will need to bear the responsibilities associated with the institutional controls is necessary. All these together may be subjected to the consideration of a full range of stakeholders so that they can be aware of the various implications of the implementation of an ER project. These discussions should take place under a well-established regulatory framework and clear allocation of responsibilities.

Accordingly to the IAEA Safety Glossary (IAEA, 2007), remediation is defined as any measures that may be carried out to reduce the radiation exposure from existing contamination of land areas through actions applied to the contamination itself (the source) or to the exposure pathways to humans. A very important element in the overall remediation concept, as defined by the IAEA, is that *complete removal of the contamination is not implied*. Needless to say, the generation and management of wastes from such projects will be heavily impacted by the extent of removal works.

The terms 'rehabilitation' and 'restoration' may also be used, but they imply that the conditions that prevailed before the contamination can be achieved again, which is not really necessary. Instead this chapter uses the term 'remediation', which does not have these implications. ITRC (2002) discusses decision-making methodologies and case studies as applied to the clean-up of radioactively contaminated sites.

8.2.1 Scoping survey

A central feature of the ER process is characterization. In this context, characterization refers to those investigations, specifically including measurements, undertaken to provide information and data about the contamination and affected site environment. Characterization steps usually taken include:

- evaluation of the severity of the problem in terms of radionuclide concentration or dose levels to determine whether there is a need to remediate;
- evaluation of the remediation alternatives including the feasibility, cost, waste generation and management, and risk reduction;

- design of the selected remediation option;
- implementation of the remediation option; and
- verification and/or monitoring of the remediation.

Characterization is a necessary prerequisite to provide critical information and data for each assessment step in this process. Multiple characterization activities are common, with each characterization activity focused on gathering the information essential for the particular type of assessment being conducted.

While the general process of dealing with a potentially contaminated site is applicable to most problems, it may result in a range of characterization activities that vary widely in terms of scope, cost and schedule. For example, a small 'hot spot' of radioactively contaminated soil resulting from a recent small spill may be surveyed, hand shovelled up into a small container for proper disposal elsewhere, and the soil replaced with clean soil in a few hours. The related characterization activities would have amounted to field survey instrument measurements of radiation prior to and after the hand shovelling.

Alternatively, the source of contamination may have been a leak of radioactive material that contaminated not only the surface soil in the immediate vicinity of the leak but also distant areas, the subsurface soils and groundwater. Migration of the contaminant might now threaten the environment and population away from the leaking source. In this instance, the components of the assessment process may be more complex and, consequently, the characterization activities may be more in number, more elaborate, and require years to complete.

Major factors to be taken into account in site characterization include:

- Characterization can be a large consumer of project resources. Mistakenly, its practical importance to solving the problem may not always be understood or appreciated. In some instances, the characterizations may be the 'last word' measurements (e.g., for peripheral areas) and, as such, their credibility is vital.
- The amount of characterization should be proportionate to the extent of the likely remediation effort. Over-characterization can result in a disproportionate fraction of the budget being spent on measurements, leaving insufficient means to carry out acceptable remediation.
- Characterization should be adequate to allow a properly designed remediation; one that does not involve excessive amounts of unnecessary effort or environmental damage.
- Characterization efforts should be sufficient to demonstrate the existence of clean areas and to provide credible assurances that un-remediated areas are safe.

- Characterizations should have a sufficiently broad focus that any other unknown contaminants are detected at a stage when they can be dealt with efficiently.
- The characterization, in the first instance, and the subsequent remediation should not make things worse by ill-advised first attempts that magnify or spread the problem. A guiding principle can be 'first, do no harm'.

The reader should note that all factors listed above are relevant to the subsequent generation and management of wastes.

Details on characterization methodologies, techniques and instruments can be found in IAEA (1998) and ITRC (2006). Practical experience, including also R&D work, can be found in IAEA (2000). It should be noted that characterization plays an essential role in the end of an ER project to certify compliance with end-state criteria and allow the planned reuse of the site. Details on post-ER characterization are given in IAEA (1999a). Figure 8.2 shows post-decontamination measurements of soil by Radon company (Russia).

8.2.2 Release criteria

Release criteria determine the radiological end-state of a remediated site and consequently the extent of remediation works and the amounts of waste generated. The IAEA have published a safety standard on release of

8.2 Post-decontamination measurements of soil by Radon company (Russia).

sites from regulatory control on termination of practices (IAEA, 2006b). Further guidance and examples are given in a Technical Document (IAEA, 2012). The safety standard represents good practice from within the IAEA member states and can be used as a guide by states when establishing their arrangements and regulations. Regulating the release of sites is a national responsibility.

The IAEA standard uses the term 'practice' to refer to any human activity that introduces additional sources of exposure or exposure pathways to people. The standard applies to cases where there is a proposal to release sites from the requirements for radiation protection of the appropriate regulatory body because practices have ceased.

The IAEA standard establishes an approach whereby target dose criteria are compared to a prospective effective dose assessment for a critical group of the public, above the pre-practice background levels, of that dose received after the site has been released for defined new uses. The dose assessed is the summed effective dose arising from the land, buildings and other sources that remain at the point of site release or licence termination.

The radiological protection principles of justification, dose limitation and optimization apply to decommissioning and are carried through to site release. The standard recommends a dose constraint for the released site of less than 300 microsieverts per year and a limit below which further dose reduction measures are unlikely to be warranted of approximately 10 microsieverts per year. The zone between 10 and 300 microsieverts per year is considered to be a zone of optimization for both restricted and unrestricted land release.

The IAEA approach provides both for cases where the release is without restrictions and for cases where the future uses of the land remain under some form of use restrictions. In the restricted case, it is possible to carry out prospective effective dose assessments with assumptions that certain sources remain under control and hence a greater degree of residual contamination can remain *in situ*. The standard recommends that should such controls fail, the effective dose should not exceed 1 millisievert per year.

The IAEA standard describes a generic approach to site release and licence termination which is expanded upon and developed within this chapter as a whole. The dose criteria for release are developed through evaluation of potential radiological consequences through all relevant exposure pathways into radionuclide release criteria (Becquerels per gram). These can be determined generically and set by the regulatory body or be developed on a case-by-case basis. Generic criteria may be more conservative because of the need to make generic assumptions in the dose assessment. The released site should be assessed for a variety of exposure scenarios including those in which material from the site is reused or circulated

outside of regulatory control. The assessment should take into account uncertainties such as those arising from sampling and analysis.

The standard describes the roles of the national government, the regulatory body and the operator in site release and licence termination. The national government should establish a legal framework under which termination of practices can occur. The regulatory body should establish detailed criteria and associated guidance, review submissions for site release, perform inspections, take actions if required and issue the licence termination once due process has been completed. The operator is responsible for safe completion of decommissioning, remediation, clean-up and licence termination processes under a specific management system. The management system should cover a process for licence termination, responsibilities, competency, calibration and maintenance of survey equipment, quality assurance, record keeping, independent assessment/auditing and non-conformance.

As an example of national approaches, the UK regulatory approach is given in the following. In the UK, licence termination is referred to as 'de-licensing'. Major nuclear facilities are licensed under the Nuclear Installations Act 1965. Prior to 2005 several examples of de-licensing occurred on parts of UK nuclear sites and for some small research facilities. In 2005, with progress towards large-scale decommissioning in the UK, the Health and Safety Executive (HSE), as the principal regulator, issued formal criteria for de-licensing (HSE, 2005). The main features are:

- De-licensing is taken to mean 'ending of the period of responsibility under the Nuclear Installations Act' and happens when the HSE gives notice in writing to the operator that in its opinion there has 'ceased to be any danger from ionizing radiations'.
- Any residual radioactivity, above natural background levels, which can be satisfactorily demonstrated to pose a risk less than one in a million per year (of the order of 10 microsieverts or less per year) for any reasonably foreseeable land use is taken to be broadly acceptable.
- Additionally, the operator should demonstrate that risk has been reduced to levels as low as reasonably achievable and should take into account the views of relevant regulators in respect of non-radiological contamination issues.
- All risks are taken to be additional to natural background levels for the area, including an allowance for impacts from authorized discharges and artificial background from worldwide sources.
- The IAEA safety guide on the application of the concept of exclusion, exemption and clearance (RS-G-1.7) (IAEA, 2004) contains radionuclide specific values that should be used to demonstrate achievement of

the risk criterion. Where these generic values are not used, a specific case-by-case risk assessment may be submitted by the operator.

- Where practicable, sources of ionizing radiation (e.g., a radiographic source) should not be within the de-licensing area at the time of de-licensing (but may be returned later where this does not require a nuclear licence). Materials which could be defined as radioactive waste (RAW) under UK legislation should not be present on the site.

The UK de-licensing regime follows the main features of the generic international arrangements suggested by the IAEA. The UK uses the lower of the range of optimization suggested by the IAEA for unrestricted use and has no arrangements for licence termination under restricted uses. Several significant parts of nuclear sites have been de-licensed using these criteria since 2005.

It is often the case that regulatory authorities take a conservative approach to release of areas after termination of practices because the decision represents a point at which formal control is relinquished. In many cases the most conservative dose criteria are used and the assessments are based upon very extensive site investigation. In the UK, for example, a 1 in a million per year risk target is used to achieve unrestricted release. For some sites with complex and extensive histories, it may not be practicable to achieve such rigorous criteria without tremendously costly clean-up and very large waste production. For these sites the concept of restricted reuse under ongoing regulatory controls less onerous than full licensing as required for the original practice should be an approach more widely employed. This may require *in-situ* waste 'disposal' authorizations or other forms of institutional control. The restricted release approach may be particularly applicable where the next use of the land is for industrial or new nuclear uses for which the potential for public exposure is limited.

If a pragmatic approach to release after termination of practices is not taken by all parties, there is potential for a 'greenfield' approach in which all physical assets are removed and the site returned to an essentially virgin state in order to enable release. In many cases it will be more appropriate to recognize the value of existing buildings, assets and infrastructure and attempt to retain these through the termination process for economic reuse.

8.2.3 Stakeholders

It is recognized that the presence of radionuclides causes fear, as radiation is not visible and its effects may only be noticeable after long periods of time. A remediation programme, therefore, has to address not only the scientific aspects of the problem, but also its societal dimension. One important aspect of the societal dimension is the communication between the

different parties having an interest in the problem, i.e. the stakeholders. Communication is often hampered by differing levels of knowledge of the subject and the specific language associated with it. In addition, there may be human values and expectations that are not shared by the different groups of stakeholders.

One cannot forget that ER of radioactively contaminated sites is also linked in people's mind to the prevailing views of different societies on nuclear power. The perception is that this technology had been historically associated with technological 'hubris', over-optimistic claims of its initial promoters, and military uses and secrecy; major accidents (e.g. Chernobyl and Fukushima), intense environmental concerns associated with RAW disposal and the stigma on communities associated with contaminated areas. There is also a widespread distrust in regulators, governments and practitioners to provide truthful information and manage risk responsibility. However, a key element that needs to be considered is that communities located very near existing nuclear power stations may hold more favourable attitudes to any new development than those who live much further away. This may be an indication than familiarity with the issue may be a positive element as people will tend to reject what they do not know. One potential avenue to explore in terms of public acceptance to remediation projects may be to share the opinions of communities living in remediated areas with those communities that will undergo a process of ER.

Involving the various stakeholders in the remediation programme will be beneficial to all parties concerned and it is advisable to involve them from an early point in the process.

Public participation in decision-making processes regarding the living environment is backed up by international agreements; one example is the Aarhus Declaration (UNECE, 1998). See the following excerpt from the Aarhus Declaration:

> *We recognize and support the crucial role played in society by environmental NGOs (non-governmental organizations) as an important channel for articulating the opinions of the environmentally concerned public. An engaged, critically aware public is essential to a healthy democracy. By helping to empower individual citizens and environmental NGOs to play an active role in environmental policy-making and awareness raising, the Aarhus Convention will promote responsible environmental citizenship and better enable all members of society to fulfill their duty, both individually and in association with others, to protect and improve the environment for the benefit of present and future generations.*

Remediation projects tend, to a large extent, to be driven by stakeholder (generally laypeople) opinions. Contrary to what is proposed by international recommendations, interested parties may wish to drive remediation projects well below clean-up levels that would be recommended if only risk

criteria were taken into account. It is not uncommon that in some occasions it is suggested/demanded by laypeople the return of the contaminated land to the conditions prior to the occurrence of contamination even if no commensurable benefit for the population/community potentially affected by the contaminated land were achieved. This tendency may cause 'over-remediation' of the site or expenditure of resources (e.g., due to excessive production of wastes) which are greater than necessary in terms of cost–benefit. In other words, resources that could be invested in other priorities, with clearer and measurable social benefit, will be spent in favour of the remediation of the site with the objective of meeting the demands of the target community. These demands may be sustained mainly by the perception and fear of radiological impacts rather than by the real effects that would be incurred by the population. A comprehensive overview of non-technical factors in an ER project (with a focus on stakeholders' views and factors) is given in IAEA (2002). In particular, this report expands on the role that planned (or preferred) land use may play in the ER decision making. It is evident that residual (post-ER) radioactive concentrations greater than 'greenfield' criteria may prevent certain uses of the site (e.g., residential); conversely, a 'brownfield' end-state may still allow reuse of the site (e.g., for industrial purposes), be acceptable to the public and cost much less. The reader could usefully consult two more IAEA reports focusing on the parallel field of decommissioning (IAEA, 2006c; 2011). The reader should note that several technical aspects are common to decommissioning and ER: Fig. 8.3 exemplifies a typical case in question, i.e. the removal of underground pipes that may have leaked and contaminated the environment.

8.3 Removal of underground pipes, Argonne National Laboratory, USA.

8.2.4 Organization and training

The organization responsible for implementing the remediation activities should have, or should have access to, competent staff to cover the following areas adequately:

- safety requirements of any permits or authorizations issued;
- regulatory standards and issues;
- radiation protection;
- conventional industrial hazards;
- data collection and evaluation;
- environmental monitoring;
- quality assurance and quality control;
- radiochemical analysis;
- geological and hydrogeological expertise;
- waste management;
- site security;
- project management (IAEA, 2006a); and
- skills and (human, technical and scientific) resources to tackle the ER challenges safely and cost-effectively.

In many cases contractors may be used to perform some or all steps of the remediation plan; however, the responsible party (licencee), as identified by the regulatory body, is required to remain responsible for the safety of all activities, including those performed by contractors. Non-radiological hazards, such as hazards due to chemical contamination, may also be present, and existing staff may not be familiar with the various aspects of the requirements for protection against these hazards. Appropriate levels of control, supervision and training should be provided to ensure the safety of workers (including contractors) with regard to all hazards.

All persons involved in the remediation should be made familiar with the contaminated area, the hazards and the safety procedures for the safe and effective performance of their duties. Specialized training may be needed in certain areas of work. For some activities, the use of mock-ups and models in training can enhance efficiency and safety. The requirements for a basic training programme and for re-training should be stated in the remediation plan.

8.3 Waste from contaminated areas: characteristics and volume

The characteristics and volumes of the wastes arising from the clean-up of a contaminated area will depend on many factors. These may be subdivided into two groups:

1. Factors affecting the radiological characteristics of the waste
   - Radioactive inventory, including amounts and physical-chemical nature
   - Geographical, topographic and hydrogeological features of the site
   - Meteorological conditions during the time environmental contamination occurred
   - Selective deposition and distribution of radionuclides
   - Decay or in-growth of radionuclides
   - The manner in which the clean-up is effected.

2. Factors affecting the quantity of waste
   - The extent, depth and nature of the contamination
   - The characteristics of the environment (prairie, desert, forest, urban, agricultural, etc.). This factor is discussed in more detail below
   - The decision on handling the affected area, i.e. stabilization of radionuclides in place, interdiction or clean-up
   - The methods used for the clean-up
   - The clean-up criteria applied, i.e. the volume of waste generated would be directly proportional to the stringency of the criteria, so that the volume will increase as the required level of residual activity decreases (IAEA, 1992)
   - The handling and packaging methods used (Fig. 8.4 shows the use of large plastic bags by RosRao company, Russia).

Rural areas include agricultural lands, wooded and grassy areas. Typical waste includes soil, organic material (crops, grass, small trees, etc.) and limited amounts of building and road material. The actual volume of waste

8.4 Use of large plastic bags for waste management by RosRao company (Russia).

will depend on the type of area and the clean-up process used. For example, if a 5 cm layer of soil and sod were removed, about 50,000 m^3 of waste could arise from each km^2. However, the volume (but not the weight) to be transported will be greater than this owing to a reduction in the density of the removed material during handling. Furthermore, additional organic waste would be produced, the volume of which will depend on the types of crop being grown. It is estimated that in certain cases an additional 50,000 m^3 of organic waste could be generated per km^2; however, the volume of the organic waste would reduce sharply as the plant material decomposed.

Urban areas could include single- and multi-family residences, commercial and industrial buildings, roads, parking areas, parks, vacant land and vehicles. Specific waste types include decontamination liquids from the clean-up of buildings, equipment, and roads, and the residues and sludge arising from the treatments. It was estimated that up to 20 m^3 of soil waste and an equal volume of vegetation could be generated from the clean-up of a garden of 200 m^2 if a 10 cm layer of soil were removed (IAEA, 1992).

8.4 Decontamination methodologies and techniques

Over the past decade, a number of remediation techniques have been developed worldwide to deal with the environmental clean-up of radioactively contaminated sites. These techniques vary in terms of sophistication and costs and must be selected on a case-by-case basis. However, the development of a successful remediation programme does not only rely on the availability of technology and expertise. Good management plans are also needed as well as appropriate communication with the various stakeholders, as pointed out in previous sections.

One important factor determining the selection of remediation technology(ies) is the area radiation levels. It is clear that excavations under low contamination/radiation conditions can be performed 'hands on', whereas at Fukushima many of the contaminated streams cannot be handled this way due to excess radioactivity. In general *in-situ* treatments (see below) entail less exposure to the workers and would be preferable in case of high contamination/radiation.

ER may face specific challenges not only because of the lack of resources but also because of the lack of appropriate technology, or the lack of experience in using new or imported technologies. These aspects altogether can end up constituting important barriers for project implementation. However, experience has shown that with appropriate planning and assistance, remedial actions are more likely to be implemented. As such the interaction of more experienced countries in ER with less experienced ones facilitated by international organizations and other donors may lead to better conditions for full implementation of projects.

This section presents particulars on ER technologies (control and treatment). The technologies addressed can be categorized as follows:

- *in-situ* treatment,
- removal of contamination; and
- *ex-situ* treatment.

Details on these technologies can be found in IAEA (1999b) and Hamby (2012).

8.4.1 *In-situ* remediation technologies

In-situ remediation technologies for control or treatment of soils and groundwater are increasingly being investigated because they offer the potential for:

- significant cost reduction of clean-up by eliminating or minimizing excavation, transportation, and disposal of waste;
- reduction of health impacts on workers and the public by minimizing exposure to waste during excavation and processing;
- significant reduction in ecological impacts, and
- remediation of inaccessible sites, including deep sub-surfaces and in, under, and around buildings.

In-situ technologies can be subdivided into five major groups:

1. Containment technologies (e.g., bottom sealing, surface capping, polymer concrete barriers, cryogenic barriers, fluidized-bed zeolite system, plasma arc glass cup, slurry wall, soil/cement wall, vitrified barriers).
2. Solidification and stabilization (e.g., lime-fly ash Pozzolan system, organic binding, Pozzolan-Portland cement system, sorption, *in-situ* encapsulation, *in-situ* compaction).
3. Physical-chemical treatment (e.g., de-chlorination, electro-acoustics, electro-kinetics, neutralization, oxidation/reduction, precipitation/flocculation, soil flushing, *in-situ* steam/air stripping, simultaneous injection, extraction and recharge, vacuum extraction).
4. Thermal treatment (e.g., radio frequency and electromagnetic heating, *in-situ* vitrification).
5. Biological treatment (e.g., biomass remediation, biodegradation).

8.4.2 Materials removal technologies

- Removal of vegetation.
- Removal of surface soil (e.g., standard or remote excavation; cryogenic removal; dust control).

8.5 Soil washing plant in Kurchatov Institute (Russia).

8.4.3 *Ex-situ* treatment technologies

- Physical processes (e.g., physical retrieval; over-packing/re-packaging/re-drumming; screening; soil washing; high gradient magnetic separation; solidification; vitrification/ceramics; incineration; filtration/ultra-filtration; reverse osmosis/membrane processes; solar evaporation); Fig. 8.5 shows a general view of the soil washing plant at Kurchatov Centre (Russia).
- Chemical processes (e.g., chemical/solvent extraction; heap leaching; enhanced soil washing; enhanced soil leaching; chemical precipitation; ion exchange; electro-dialysis; adsorption; aeration).
- Biological processes.

8.5 Waste transportation

During the clean-up of very large contaminated areas, the loading and transportation of much of the wastes to the disposal site could probably be accomplished using conventional earth-moving equipment from the construction industry. Some modifications may be beneficial, such as the addition of shielding between the driver's cab and the box of the dump truck. If the disposal site is located within the clean-up area, much larger equipment than that used on the site in major civil engineering and mineral extraction projects could be used. Large volumes of contaminated soil, concrete, asphalt, equipment, vegetation, etc., could arise from the clean-up

of a large contaminated area. The removal of a thin (average thickness of about 5 cm) layer of contaminated material from a 7 km radius around a damaged facility could result in 8×10^6 m^3 of waste which has to be transported to a disposal site and buried. The loading and moving of such large volumes of soil is time-consuming and expensive, but the experience is not unique (e.g., for the construction of large earth dams, or in mining).

The loading of the contaminated soil could be done:

- using equipment such as wheeled or tracked loaders and excavator loaders with capacities of 30 m^3 or more;
- using a force feed loader with a conveyor which can pick up a layer of soil or soil from large heaps and dump it directly onto a truck;
- using vacuum pickup systems for certain types of soil under dry conditions.

Water spraying equipment, to dampen soils during handling under very dry conditions, may be useful to minimize dust production. Highly contaminated soil may have to be sealed in appropriate containers for transport.

The contaminated wastes could be transported using one or more of the following techniques:

1. Moving the layer of contaminated soil directly into depressions or specially excavated trenches using scrapers, bulldozers or graders.
2. Loading the soil into dump trucks for transport to the disposal site.
3. Loading the soil into railway cars for transport to the disposal site. The choice of rail transport depends on the availability of railway lines in the vicinity of the clean-up and disposal sites. The economic factor in the decision may be offset by the fact that rail transport results in smaller radiation exposure to transportation workers and involves less interaction with the public than does truck transport (IAEA, 1992).

8.6 Waste disposal

The objective of disposing of radioactive wastes is to confine the radionuclides within the repository site until they no longer represent an unacceptable risk to the environment and the public. A repository should fulfil two important and related functions in this regard: (1) to limit dispersion of the radionuclides contained in the wastes by waterborne and airborne pathways, and (2) to protect the waste from surface and near-surface deteriorating processes such as erosion or intrusion by humans, burrowing animals or deep-rooted vegetation.

The radionuclides of longer term concern in the soil after an accident at a nuclear power plant are ^{90}Sr and ^{137}Cs, both with a half-life of

approximately 30 years. After about 300 years, the concentrations of these radionuclides in soil would be about 0.1% of the concentrations immediately after the accident. Therefore, a storage facility capable of containing these wastes for several hundred years should be suitable for most of the soils collected.

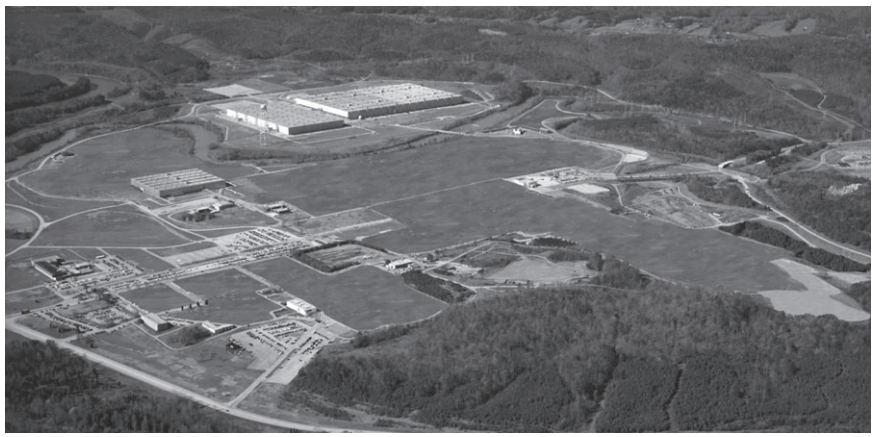
The type of facility selected for disposal of the soil will be dictated by many factors, including the availability of equipment to move the wastes, the volumes to be moved, the distances involved, the availability of natural or man-made disposal sites such as quarries, mines or depressions, and the hydrogeology and geology of the area. The basic factors which must be considered in order to achieve a suitable disposal repository system are: the quantity and nature of the wastes, the engineering features incorporated into the repository design, the site characteristics and the time period allowed for institutional control. It is likely that for transport of large quantities of material, haulage cost will be the largest component of the overall cost.

Conditions are combined in the safety assessment to achieve a disposal system that will meet the regulatory or desired environmental protection requirements. For example, a special cover to prevent intrusion by humans would not be required if the institutional control period is expected to be longer than the hazardous life of the wastes.

8.7 Future trends

ER technologies are adequate in many cases. The most obvious intervention is the simple removal of contaminated material. However, this may lead to a high cost of contaminant removal and treatment and storage of removed material. The challenge is to find reliable methods which minimize the amount of contaminated material to be removed or to allow the contaminated material to remain on-site, without major impact on the planned land use. Figs 8.6(a) and (b) provide an aerial view of the East Tennessee Technological Park, USA (ETTP or K-25) in its operational state and after remediation and reindustrialization, respectively. Contaminant migration to areas surrounding a pollution source is a major environmental concern and methods are needed to control the spreading of pollution. These technologies could provide short-term containment while the polluted volume of soil (sometimes called plume) is being remediated or long-term containment for sites presenting no immediate danger. High priority is placed on treating plumes *in situ*, so that potential worker and public exposure is eliminated. *In-situ* methods minimize waste material and reduce costs. Biological remediation systems utilize the natural ability of plants or microbes to metabolize, absorb, oxidize or reduce radioactive compounds, and may produce significant cost savings.

(a)



(b)

8.6 (a) ETTP in operational state; (b) ETTP after environmental remediation and re-industrialization.

Improved methods of constructing impermeable barriers at greater depths and new methods of installing subsurface containment barriers are necessary to reduce construction costs and increase their depth of application (Laraia, 2011).

Regardless of progress gained over the last 10–15 year – mostly as the result of opening up of countries to international co-operation and assistance – several issues remain to be solved, or even recognized, in less developed countries. The following is a list of problematic areas where further efforts are needed on the part of the international community.

8.7.1 International assistance

It has been argued on many occasions that despite resources being made available to some countries, little improvement has been observed. Is there a need for better coordination between the organizations involved? Should international donors improve their interactions with recipient countries to optimize the outcomes? Are there indicators available to measure the effectiveness of the aid given to the recipient countries in terms of concrete achievements? How do the countries manage to make the best use of the support they receive? What are the constraints besides lack of funds: availability of local human resources? Technical capability at the individual and institutional levels? Conflicting legislation? Political issues? Cultural environment? Can financing mechanisms be improved?

8.7.2 Safety and regulatory issues

One of the points that should be explored is to what extent a regulatory framework is of utmost importance for the implementation of ER programmes. But this is not enough. On top of that, regulatory requirements must be well understood by all the sides involved, something that is especially challenging when one takes into account the philosophical elements embodied in the radiation protection principles. Despite international recommendations, final, mandatory decisions are taken in political and judiciary environments that do not necessarily possess the proper technical background, often leading to total removal of the contamination and excessive (and unnecessary) expenditures (greenfield rather than brownfield and redevelopment). It may be useful to discuss the regulatory differences in different countries (on the basis of economics or social-cultural-political environments). How best to transfer the experience from one country to another? How to establish a better flow of information between scientific community, regulators and industries? Are the industries aware of, and do they possess, a good understanding of the rationale behind the regulatory requirements? How do industries perceive the existing regulatory framework? How to improve co-operation between regulators and other players? To what extent should international guidance (e.g., from the IAEA) be tailored to individual countries? How much flexibility should be allowed for reference doses? Prescriptive (e.g., radioactive concentrations) v. non-prescriptive (e.g., cost–benefit analyses) approaches is a crucial issue.

8.7.3 Technologies in ER programmes

What technologies have proved to be effective? What needs to be improved? What are the innovations and how promising are they? Is there room or

need for extensive R&D programmes? How willing are international or local contractors to use innovative technologies? What are the risks? The business factor can be measured in terms of inexpensive vs costly technologies (e.g., *in-situ* measurements vs. laboratory analysis). How to optimize the relationship between costs and accuracy of measurements? ER planning depends heavily on predictions about the behaviour of pollutants in different compartments (soil, groundwater, etc.) as well as on the design of remediation solutions. What are the challenges in mathematical models? How effectively and wisely are these being used? How to best educate and give training to potential modellers? What role does statistics play in pre- and post-clean-up characterization in reducing costs while ensuring safety?

8.7.4 Planning factors

Planning factors such as ER under a life-cycle perspective and non-technical issues are increasingly influential factors. Issues to be mentioned are the resources to aid good planning (with economics taken into account). How effectively are they being used? Again, how the experience from more advanced countries in the field of ER can be better transferred to less advanced ones? How to best incorporate ER in the whole life cycle of an operation and also how to optimize remediation programmes taking into account the life cycle of the projects? What are the best ways to engage stakeholders in the decision-making process? What should be communicated and how? What are the challenges in the different geographical regions of the world? How to clearly state to the public (and be convincing on it) that remediation does not mean returning the environment to background levels; instead, new productive uses can be envisaged after ER? Who are the relevant stakeholders and how to best approach them? Ethics of ER remains crucial: will optimization justify higher expenditures in affluent countries in comparison to less developed countries?

8.8 Conclusion

In the past, many nuclear activities were developed without proper consideration of environmental issues. Operations took place without established or well-addressed environmental laws and regulations. Through lack of good operating practices, contaminated sites have been created in many countries. Several contaminated sites have also been created by nuclear and radiological accidents.

Contaminated sites can ultimately lead to undesired health effects to the local residents. Environmental remediation strives to reduce the radiation exposure from contamination of land or other polluted media, such as surface water or groundwater.

In recent years a dramatic change in vision occurred: awakening awareness of environmental long-term problems has been bringing forth a move away from treating environmental problems only after they have occurred (typically at the end of service life of a facility or site). The current vision is to prevent environmental impacts from the beginning in the life cycle of a facility or activity. This life-cycle management aims to treat each stage of an operation not as an isolated event, but as one phase in its overall life. Thus, the planning covers not only each stage, but is a continuing activity, taking into account actual and projected developments. By implementing the elements of this vision, it is expected that the generation of contaminated sites as well as the need for expensive remediation programmes will be minimized.

National institutions need timely and accurate information on available remediation strategies and technologies, management options as well as guidance in dealing with non-technical factors, e.g., communications and stakeholder involvement. To resolve environmental liabilities and to avoid the generation of new contaminated sites, the IAEA and other international organizations help countries to adopt appropriate practices.

8.9 Sources of further information

Conferences dealing with radioactive waste management or decommissioning often include sessions devoted to ER. In this regard, the reader is invited to consult the series of conferences known as ICEM (International Conference on Environmental Remediation and Radioactive Waste Management, see Track 4 under http://asmeconferences.org/icem2013/) or Waste Management (WM) Symposia (http://www.wmsym.org/). In addition to the reports and papers mentioned below, the reader is also invited to consult the following comprehensive material:

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9

Safety and risk assessment of radioactive waste (RAW) and contaminated sites

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Abstract: The axiomatic basics of quantitative safety/risk assessments are discussed. Deterministic and probabilistic analysis methods are then introduced. As an illustrative example, safety assessment for the environment is given in terms of the probability of radionuclide escape from a near-surface disposal facility. Emergency accident levels are correlated with the probabilities of those accidents occurring.

Key words: safety, risk, equipment failure, probability of failure, emergent event tree, failure trees.

9.1 Introduction

Increased attention is being given to issues of safety in the nuclear field as evidenced by the large numbers of publications on this topic. These can be divided into two types according to how the term 'safety' is interpreted. The first, the subjective type, concerns the safety of the environment, population or personnel in the proximity of radioactive materials and ionizing radiation sources. The second, the objective type, discusses the safety of nuclear power plants (NPP) or nuclear hazardous objects, although the influence of these objects on the environment or people is also discussed. In the first case, safety is treated in terms of the ability to protect from the effects of ionizing radiation. In the second case, safety is treated as a property of an object, i.e. the property of not rendering an action hazardous, not resulting in contamination or not resulting in the spread of radioactivity. This ambiguity can lead to misunderstandings in safety assessments. For example, with respect to the ability to protect, it is necessary to take into account the availability/absence of radiation detectors and means of personal protection. If, on the other hand, safety is the property of an object and the need is to avoid hazardous action, taking, for example, the NPP, then other factors must be included associated with the object's composition, for example the physical barriers designed into the system.

Scientific safety assessment requires that the true character of both subjective and objective safety problems is considered quantitatively in mathematical terms. In this chapter safety assessment is considered using the following system of definitions and axioms:

Definition 1. The safety of an object indicates the state of its immunity from the harmful influence of other objects or factors dangerous to it.

Immunity assumes the ability to resist harmful influence. This ability can be realized through the presence of systems and the elements which prevent penetration of the dangerous object and characteristics of the protected object, i.e. through the presence of a specified protective shield separating the dangerous and protected objects. In other words, it is possible to say that the object is protected if it is supplied with a protective shield (Fig. 9.1).

Definition 2. The protective shield is the means of reducing or eliminating harmful influence on the protected object.

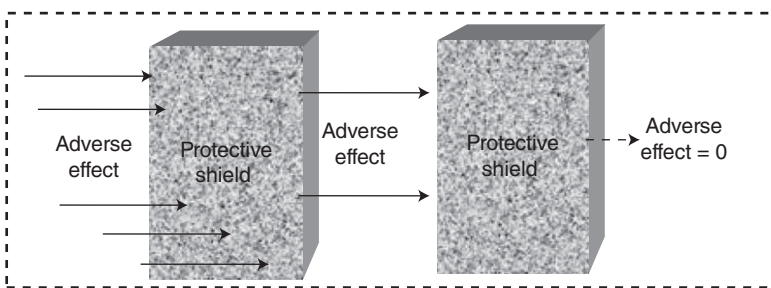
Objects may be divided into the protected or dangerous by analyzing their interaction. The object which has harmful influence is considered as dangerous. The object, whose safety should be ensured, is protected. For the theory to be developed we must formulate:

Axiom 1. There are protected objects and dangerous objects or factors.

This axiom is needed to clearly establish the safety of the object in question, as it is protected, and dangerous objects may change their roles. For example, if we consider the harmful affects of radioactive wastes (RAW) on the environment, the latter is protected and the wastes are a dangerous object. If we are talking about external influences on disposal wastes in a facility, then all the potential, unauthorized entrants such as terrorists, meteorites, infiltrating moisture, insects and rodents, that is, in fact, the environment, become a dangerous object, and the wastes are what must be protected. This shows that to distinguish the protected objects from the dangerous it is necessary to consider the postulate of their interaction:

Axiom 2. A dangerous object has a harmful influence on the protected.

From Definition 2 and the axioms, the obvious conclusion can be formulated:



9.1 Protective shield.

Safety principle: For the safety of a protected object it should be separated from the dangerous object or factors by means of a protective shield.

These definitions, axioms and the safety principle are the axiomatic basis of the safety theory as a scientific area. The safety principle enables us to understand that quantitative safety assessments can be made, characterizing the state of the protective shield, its performance and reliability. Therefore, the subject of study of the safety theory is the protective shield. The theory involves the use of mathematical modeling of the system's functioning as the protective shield and the components that make up the protective shield, as well as numerical methods and natural, physical experiments to determine the characteristics and parameters of these systems.

9.2 Deterministic and probabilistic analysis methods

The essence of deterministic safety analysis (DSA) is the solution of differential equations simulating the processes of radionuclide transfer and ionizing radiation from the source to the environment. Solving the equations gives the concentration or the volume activity of certain radionuclides at a given distance from the source at a given time. Comparing these results with regulatory requirements enables conclusions to be drawn about the safety of the environment. At the next stage of the analysis, calculated concentrations are transferred to the projected radiation doses to control groups of the population, using different scenarios and pathways of radionuclides in the body. Comparing the predicted and normative values enables conclusions to be drawn about the safety of the population. The equations used contain deterministic functions and coefficients, so giving the name to the method.

Solving the equations requires many simplifying basic assumptions including: use of a point source, the uniformity of the environment, fixing in space and time the parameters and coefficients of the equation and baseline data. In the case of an extended source, such as a near-surface disposal facility, account must be taken of its heterogeneity. To solve the equations, depending on the model, 10–16 parameters must be set that describe the source properties, the engineered barriers and the surrounding geological environment. These settings are heterogeneous in the physical sense, and so difficult to define and with a large scatter in numerical values. Equations are solved by the usual finite difference method using ready-made software products such as MathCard Enterprise Editoria V11.A, AMBER from Quantisci (UK), ModFlow (USA), or MT3D (USA).

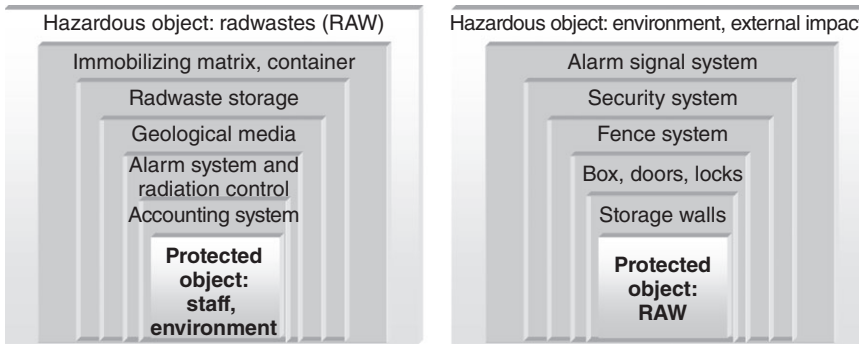
Many parameters in the equations vary greatly in both space and time. This is a consequence of the stochastic nature of the environment and the changing external conditions. Therefore, solutions to the equations must be

a random variable. It is well known empirically that the concentration of radionuclides in different, even neighboring areas, is substantially different. Such a property of the distribution of radioactive contamination is particularly pronounced after an accident with a significant release of radioactive substances into the environment such as at Chernobyl or Fukushima. Once an accident occurs, the migration of radionuclides is governed by largely random processes, so it is natural to use the methods of probabilistic safety analysis (PSA). The purpose of PSA is to estimate the probabilities of certain accident scenarios over a given period of time and to identify the weakest elements of the complex in the disposal of RAW. The basis of the PSA methodology is a systematic analysis of the radiation-dangerous object, the selection of systems and components that make up the protective shield, and making event tree and failure trees with subsequent calculation of the probabilities of various scenarios of accident events. To perform the calculations it is necessary to access fundamental homogeneous data on the physical properties of elements of the physical barriers to ensure retention of radionuclides in the bulk of the medium. This data includes the following interdependent and replaceable parameters: the intensity of the element failures, mean time between failures, and failure probability. Such information can be obtained from technical regulations, manuals and handbooks, as well as the results of physical and field experiments, mathematical modeling or calculations by deterministic models.

9.3 Safety and risk assessment

The key subject of study in the safety theory is a protective shield and this can be quantified by evaluating its reliability and efficiency. To do this, we need to establish what constitutes a protective shield – which systems and components. Considering the situation where the protected object is the environment, and the dangerous objective RAW, the environment provides the multi-barrier protection system, as shown in Fig. 9.2(a). In addition to the engineered barriers, the protective shield must contain a system for providing information – or radiation monitoring. The purpose of a monitoring or alarm system is to provide timely and reliable information on the dangerous object or radiation and ecological state of the environment.

The case where the protected object is the RAW, and the hazardous factors are the impact on them from the environment, is illustrated in Fig. 9.2(b). The physical system of barriers may remain the same, but in a functional sense, these barriers are quite different objects. Indeed, in the first case, the barriers are characterized by parameters that reflect the migration of radionuclides, such as diffusion factors, absorption, porosity, filtration, mean time between failures, probability of the nuclides release, etc. In the second case, these same barriers are described by a different set of



9.2 Protective shield for (a) environment and (b) radioactive waste.

parameters, including, for example, seismic stability, resistance to impact and thermal loads, and the probability of failure under the influence of these factors.

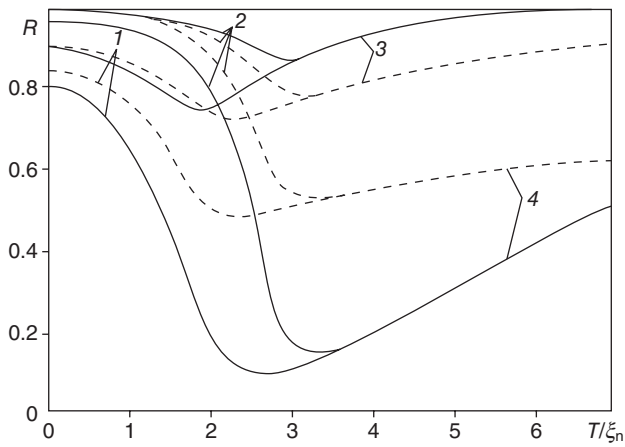
Thus the state of engineering and physical barriers, i.e. the physical part of the protective shield, can be evaluated quantitatively by the probability of failure-free operation. The larger this probability, the higher the safety. For this purpose, well-developed methods of reliability theory are used. The effectiveness of the information part of the protective shield can be evaluated using the information theory. To quantitatively assess the reliability of the information part of the protective shield, the notion of risk must be introduced.

The problem of risk is part of the safety theory. The complexity of the problem arises because to date there has been no universally accepted concept of risk. There are many interpretations and approaches to risk assessment (Petrin *et al*., 2003). The most commonly identified risk is associated with the probability of adverse outcome, the damage from the consequences of accidents or natural disasters and with the frequency or flow of the emergent events. The situation reached such a state that one of the publications (ICRP, 1991) was invited to use the word 'risk' not as a defined term, but as a metaphor, so not giving it a specific scientific meaning. One approach to a constructive solution to this problem is to attribute the concept of risk to loss of information about the state of the controlled object (Puzanov, 1992, 1993, 2005, 2010). The more information about the current state of the controlled object we have, the less the risk of undesirable consequences. Knowledge about the state of the object comes from the information part of the protective shield via the monitor signals. If we assume that risk arises from the loss of this information, the risk assessment can be quantified using the signal detection theory. Correctly and timely received

signals about the state of the object can prevent an accident, or at least minimize undesirable consequences, so reducing the risk.

The information part of the protective shield and the controlled object, regardless of whether it is dangerous or protected, can be integrated from a functional point of view. This complex is characterized by a set of physical parameters, as well as a set of the physical barriers. These parameters are: the variance of the logarithmic amplitude of the signals ξ_s^2, the variance of the noise ξ_n^2, the signal detection threshold T, the expected signal flow λ_s, the nominal (modal) of the signal amplitude E_0, the expected signal/noise ratio $\chi_0 = E_0/\xi_n$, the realized signal/noise ratio $\chi_0 \leq E_s/\xi_n$, the signals detection band β, and the dead-time of the information system τ. Owning these parameters, we can construct a self-similar risk characteristic $R \equiv R(\xi_s, \chi_0, \chi, \lambda_s/\beta, \beta\tau, T/\xi_n)$ as a function of six dimensionless variables. As an example, Fig. 9.3 shows the risk characteristics of a hypothetical information system. The graphic shows that the risk alternates with the alteration of the threshold T. If $T < T_0$, the alarm system works very frequently, an operator has no time to react to the signals, or he stops paying attention to it, so the risk increases. If $T > T_0$ then the alarm system is less sensitive and the signals occur more rarely. If a useful signal about the accident state of the object arises, it may be missed, so the risk increases again. If the threshold is optimal, $T = T_0$, risk is minimum.

One of the main issues in risk theory is the search for such parameters of the information system at which the risk reaches a minimum. It has now been established (Puzanov, 2010), that the local risk minimum is achieved when the condition reaches:



9.3 Risk characteristic: $\lambda_s/\beta = 0.1$ (1) and 0.01 (2); $\chi_0 = 2$ (3) and 7 (4); $\xi_s = 0.5$ (—) and 2.3 (----); $\beta\tau = 1$; $\chi = 3$.

$$\left(\frac{\lambda_s}{\beta}\right)_{opt} = \exp\left(-\frac{T_0^2}{2\xi_s^2}\right), \quad [9.1]$$

where the left-hand side of the formula is the optimal ratio between the expected flow of useful signals and their detection band, and in the right-hand side the symbol T_0 is the optimal detection threshold of the signals.

The approach developed to solving the problem of risk is based solely on the analysis of the complex 'object-control system' as a whole, without involvement of extra-systemic socio-economic and hygiene categories.

9.4 Application to the case of radionuclide escape from a near-surface disposal facility

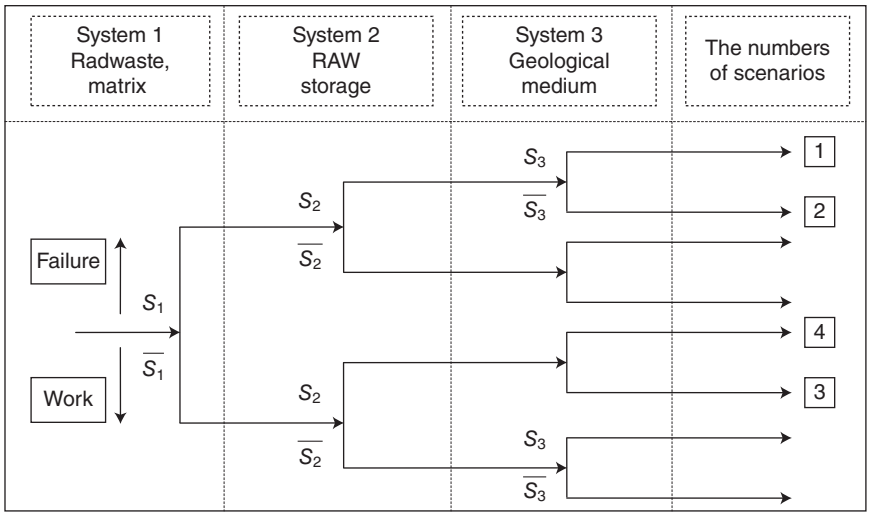
This approach to safety assessment is demonstrated using an example with the environment as a protected object. The dangerous object is the RAW contained in a near-surface disposal facility. Initially, we need to identify which engineering and physical barriers prevent escape of radio nuclides into the environment. In the case of processing plants, barriers include: piping, valves, pumps, physical or radiological protection, etc. In the case of the disposal facility, the multi-barrier protective shield consists of three systems:

1. the matrix (container), which contains the RAW,
2. the repository, as an engineering and construction structure, and
3. the geological environment in which the repository is located.

To formalize the numerical calculations, the functioning of the individual protection system is described by the binary relation of 'failure' to 'work'. Decomposition of the protective shield to the individual systems is an essential part of the safety analysis. An emergency event which may be an incident, accident, occurrence, or situation, etc., is defined by a sequence of failures of the systems and components leading to adverse effects such as a loss of control over the ionizing radiation sources or uncontrolled escape of energy and matter from a dangerous object into the environment. This sequence of events is called a scenario. If a complex of the protective shield is divided into N systems, there are only $M = 2^N$ scenarios. In the case of near-surface disposal facility, $N = 3$ and $M = 8$. Functioning of the complex is displayed graphically in Fig. 9.4. Such a diagram is called an event tree.

In this diagram, the systems are presented in the form of columns. The rows represent the state of these systems. Armed with the event tree, it is easy to calculate the probability of each scenario from the general formula:

$$P_m = \prod_{n=1}^{N} S_n^{(1-q_{nm})} \overline{S}_n^{q_{nm}}, \quad [9.2]$$



9.4 Emergent event tree for activity of disposal complex.

where S_n, $n = 1, 2, \ldots, N$, is the failure probability of the n-th system, and $\overline{S}_n = 1 - S_n$ is the probability of the work state of this system; $m = 1, 2, \ldots, M$ is the scenario number; q_{nm} is the state indicator of the system; $q_{nm} = 0$ if in scenario m the system n is in a failure state, and $q_{nm} = 1$ if the system is in a working state.

The probabilities S_n are calculated by analysis of the failure trees for each system. The failure tree is a logical connection between the system elements, connected by symbols 'OR', 'AND', corresponding to addition or multiplication of the random failure events. The symbol 'OR' links together the group of elements, failure of at least one of which leads to failure of the entire group. The failure probability of such a group is calculated from:

$$S_{Jor} = 1 - \prod_{j=1}^{Jor} (1 - E_j), \qquad [9.3]$$

where E_j is the failure probability of the j-th element from the group; J_{or} is the number of elements in the group. The symbol 'AND' combines the group of elements, only the joint failure of which leads to failure of the whole group. The failure probability of such a group is calculated from:

$$S_{Jand} = \prod_{j=1}^{Jand} E_j, \qquad [9.4]$$

where J_{and} is the number of elements in the group.
The failure probability of elements is calculated from:

$$E_j = 1 - \exp(-\lambda_j t), \qquad [9.5]$$

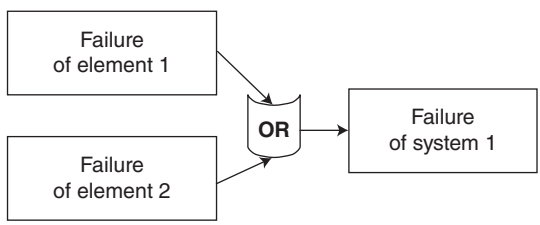
where λ_j is the failure rate of a given element; and t is the time from start of observation or operation of the disposal facility.

We now consider analysis of alarm events associated with possible escape of radionuclides into the environment from the complex for disposal of RAW. Since the complex consists of three systems, event three is the same as in Fig. 9.4. Since not all scenarios can be realized, we consider only the four scenarios of the alarm events that have physical meaning:

- Scenario 1. Failure of all systems that make up the disposal complex.
- Scenario 2. Joint failure of the matrices and the disposal facility.
- Scenario 3. Only failure of system 2 (the repository). Physical representation of this scenario is to destroy the structural elements of the repository with probable leakage of radionuclides beyond.
- Scenario 4. Only joint failure of systems 2 (the repository) and 3 (the geological environment). Physical representation of this scenario is the escape of radionuclides from the disposal facility and their migration into the geological environment.

As the scenarios are interdependent, the sum of probabilities of all scenarios is 1. Therefore the probability of accident $P_{ac} = P_1 + P_2 + P_3 + P_4$ and the probability of the work $P_{work} = 1 - P_{ac}$.

In the model representation, system 1 consists of two elements. Element 1 is the RAW itself, contained in a matrix or container. Element 2 is the body of the matrix. System 1 failure occurs when a failure occurs in element 1 or element 2 or both. Hence, these two elements are working on an 'OR' scheme. The relevant failure tree is shown in Fig. 9.5. Physical representation of element 1 failure is the radionuclide escape from the matrix body as a result of diffusion and leaching. Physical representation of the element 2 failure is the matrix degradation during its aging, corrosion, and cracking. Although the physical processes of failure of these two elements are interrelated, from a model point of view it is convenient to present them as independent. Conditionally we can accept the failure rate of element 1 as

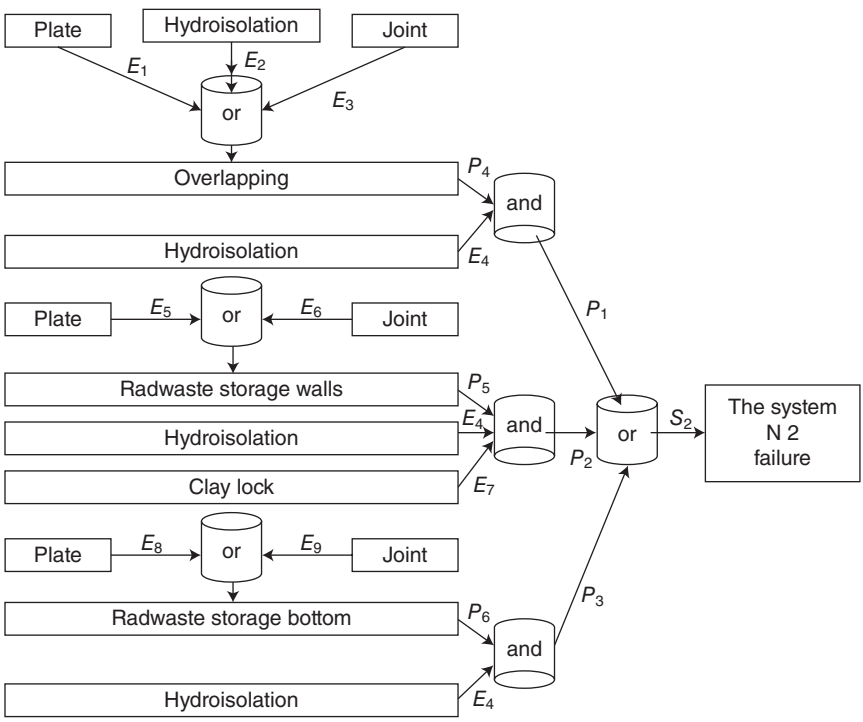


9.5 Failure tree of matrix with radioactive waste.

$\lambda_1 = 9.1 \times 10^{-11}$ 1/year and of element 2 as $\lambda_2 = 1.6 \times 10^{-8}$ 1/year. From Eq. [9.5] we calculate the failure probability of elements 1 and 2. Then the failure probability of system 1 can be calculated from Eq. [9.3].

We now discuss the physical barrier in the RAW repository (system 2). From the system analysis standpoint, this system can be regarded as consisting of the following elements: covering slabs, walls, a bottom, waterproofing. In turn, these elements may be composed of elementary units: concrete slabs, cement joints, beams, etc. The failure tree of system 2 is shown in Fig. 9.6.

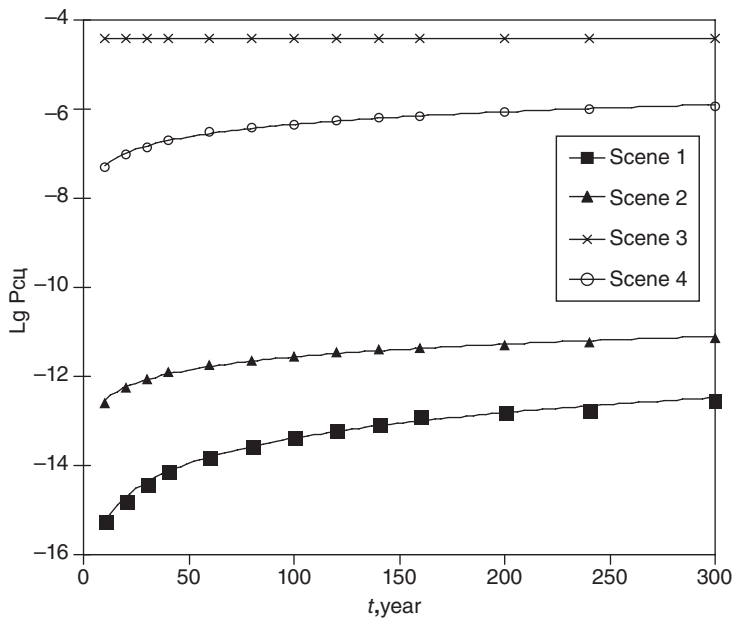
It should be noted that, depending on whether the composition of a structural element contains slabs or joints, they may have different performance parameters. Therefore in Fig. 9.6 the same names of the elements are presented with different numbers. The logical scheme of calculation can be easily understood from Fig. 9.6. System 2 failure occurs when the events are implemented by the probabilities denoted in Fig. 9.6 by symbols P_1, P_2 and P_3. Then the system 2 failure probability is calculated from formula: $S_2 = 1 - (1 - P_1)(1 - P_2)(1 - P_3)$ in accordance with Eq. [9.3]. Probabilities P_1, P_2, and P_3 are calculated using Eq. [9.4]: $P_1 = P_4E_4$, $P_2 = P_5E_4\ E_7$, $P_3 = P_6E_4$.



9.6 Failure tree of repository.

Probabilities P_4, P_5, and P_6 are calculated in accordance with Eq. [9.3]: $P_4 = 1 - (1 - E_1)(1 - E_2)(1 - E_3)$, $P_5 = 1 - (1 - E_5)(1 - E_6)$ and $P_6 = 1 - (1 - E_8)(1 - E_9)$. Probabilities E_j are calculated from Eq. [9.5].

Physical representation of the system 3 failure is the escape of radionuclides beyond the sanitary protection zone, or their penetration into the aquifer. From the standpoint of the system analysis, the geological environment is composed of two elements. The first element prevents the horizontal migration of radio nuclides on their way to the border of the sanitary protective zone. The second element retains radionuclides during their vertical migration towards the aquifer. Obviously, these two elements are working on an 'OR' scheme, so that the system 3 failure tree is the same as in Fig. 9.5. The failure rate of these elements is assumed to be equal to the reciprocal of mean time between failures. This is the radionuclide migration time, which may be calculated from known geometric data and the speed of horizontal and vertical migration. Migration rate can also be obtained from the results of field measurements or from calculations by deterministic models of the radionuclides transport. With this information, Eq. [9.3] calculates the system 3 failure probability. Then Eq. [9.2] is used to calculate the probability of each scenario. As an illustration, the results of such calculations are shown in Fig. 9.7. On the graph, ordinate is logarithm of the scenario probability, and the horizontal axis gives the time period during which this scenario can be realized.



9.7 Failure probability of disposal complex.

9.5 Correlation of emergency accident levels with probabilities of occurrence: implications for the safe operation of facilities

Any technological installation, as well as the disposal facility of RAW, is a source of potential danger because in the course of operation, they are forced to interact with the environment and, therefore, to provide some impact on it. Adverse effects associated with contamination of the surrounding area can be assessed via the radiological condition of the territory using a five-score system: (1) normal; (2) satisfactory; (3) accident; (4) emergency; (5) disaster (Busygin *et al.*, 2009). The ratio $r = A/RL$ can be used for quantitative assessment. Parameter A is an actual level of contamination, and parameter RL is the reference level, typical for this area. The value of r is continuous and varies widely, which complicates the classification and interpretation of the effects of radioactive contamination. Therefore, we propose a system assessment area, based on rankings of r in accordance with Table 9.1.

Contamination of the environment and the site can clearly result from accidental events in radiation-hazardous facilities. Regulatory and legislative documents allow *a posteriori* estimation and classification of emergency events, based on measurements of actual contamination levels and comparing these results with a certain threshold. However, in practice it is necessary to calculate and predict the consequence of events prior to their occurrence, i.e. to give an *a priori* assessment of the events. Since the events themselves, as well as their effects, are effectively random, then the evaluation must be made in terms of random variables, i.e. must have a probabilistic nature. An international scale is used to link the seven levels of technological accidents at NPP and their consequences on the environment (INES, 2008) as given in the first two columns of Table 9.2. Based on this scale, we propose an additional relationship between the levels of incidents with their probabilities for all radiation-dangerous objects, which do not belong to the nuclear fuel cycle facilities. These relationships are given in columns 3 and 4 of Table 9.2 (Puzanov *et al.*, 2004, p. 220).

In some regulations (GAN, 2000) for objects which do not involve the nuclear fuel cycle, a three-score grading system for the class of incidents and

Table 9.1 Score system for the assessment of the state of the site

Range of value r	$r < 2$	$2 \le r < 4$	$4 \le r < 6$	$6 \le r < 9$	$r \ge 9$
Ball	1	2	3	4	5
Site state	Normal	Satisfactory	Abnormal	Emergency	Disastrous

Table 9.2 Levels and probabilities of incident

Consequences	Trouble-free operation probability	Probability should not exceed	Incident level
Non-essential difficulties in operation	0.80	0.20	I
Essential difficulties in operation	0.90	0.10	II
Short-term stop of equipment	0.95	0.05	III
Stop of equipment at large material losses	0.99	0.01	IV
Complete destruction of construction	0.999	0.001	V
Destruction accompanied with danger for people's health	0.9999	0.0001	VI
Disastrous destruction accompanied with a lot of victims	0.99999	0.00001	VII

their consequences is recommended, as well as liaison on the levels of contamination. Using Table 9.3 we can associate a class of incident with the probability of their realization and we can specify the requirements for safe operation of facilities. As a case study, we consider the emergency situation at the NPP 'Fukushima-1'. According to many experts, the situation is consistent with a IV–V level of complexity. Initially the reactor coolant system failed under exposure to the earthquake measuring 9 on the Richter scale. According to Table 9.3, we can conclude that, due to this level of earthquake, the failure probability of the cooling system must not exceed 10^{-3}–10^{-2}. It is important to emphasize that the initiating event, i.e. earthquake itself, is not included in the script, since it is not an element of the event tree but is the external condition under which the event tree is realized.

9.6 Sources of further information

1. Safety indicators in different time frames for the safety assessment of underground radioactive waste repositories. IAEA-TECDOC-767, 1994.

   Among other safety indicators, the notion of risk is introduced. Three scenarios of the release of radionuclides ranging from disposal facility to the individual are shown. The hybrid safety indicator is suggested in the form of the product of the scenario probability on the effective dose assumed in the result of this scenario.
2. Hossain S. Safety assessment for near-surface disposal. First workshop on RW management infrastructure. IAEA, Vienna, 18–20 April 1995. A brief guide for safety assessment is given.

Table 9.3 Generalized indicators of the ecological impact on site in result of radiation accident on various level damages

Emergency class (GAN, 2000)	Dose rate at distance 1 m above surface D, Gr/day (GAN, 2000)	Emergency level in international scale (INES, 2008)	Emergency probability must not exceed	Dose rate D, Gr/day (Busygin *et al.*, 2009)	Ecological damage (Busygin *et al.*, 2009)
Loss of control at radiation source which may be able to bring about pollution of environment, but not bring it about	$D < \mathrm{RL}$	I, II	0.1	$D \leq 10^{-6}$	Be absent
Loss of control at source with consequent pollution of environment exceeding the reference level, but not exceeding legislative norms	$\mathrm{RL} \leq D \leq 2.4 \times 10^{-3}$	III, IV	0.01	$10^{-6} \leq D \leq 10^{-4}$	Chromosomal divergence by animals (α, β, γ-radiations) and plants (β, γ-radiations)
Loss of control at source with consequent pollution of environment exceeding legislative norms	$D > 2.4 \times 10^{-3}$	V, VI, VII	0.001	$10^{-4} \leq D \leq 10^{-2}$	Mutations by animals and plants
				$10^{-2} \leq D \leq 10^{0}$	Missing sensible forms
				$D > 10^{0}$	Pauperization and degradation of communities

3. Safety assessment of near surface radioactive waste disposal facilities: model inter-comparison using simple hypothetical data (Test case 1). IAEA-TECDOC-846, Vienna, 1995.
   The most likely ways for the release of radionuclides from disposal to the individual are considered. Deterministic safety analysis is used and results of calculations are given for the facilities in some countries.
4. ISAM scenario generation and justification working group, working document ISAM/SGWG/WD01. IAEA, Vienna, 2001.
   The methodology of scenario development for safety assessment is considered including the lessons learnt by the ISAM group.
5. ISAM 'Radon'-type facility safety case working group, working document ISAM/SCWG/WD02. IAEA, Vienna, 2001.
   The deterministic safety analysis is given for near-surface disposal facility. Specific results of calculations are presented for radionuclide concentrations.
6. ISAM borehole safety case working group, working document ISAM/SCWG/WD03. IAEA, Vienna, 2001.
   The mathematical model of the radionuclides release from a near-surface disposal facility is presented. The deterministic analysis of safety is employed.

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10

Russia: experience of radioactive waste (RAW) management and contaminated site clean-up

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Abstract: Global challenges in contemporary development of the Russian Federation the creation of a reliable state energy power system. The use of nuclear power, new nuclear technologies, sources of energy, medical innovations require further large-scale development of radioactive materials. However, this development is restrained by the problem of radioactive wastes accumulated from the early Russian nuclear programmes. This chapter describes current activity in the sphere of radioactive waste management, including consequences of technological incidents in the Russian Federation.

Key words: radioactive waste, contaminated region, radiation techniques, spent ionizing sources, repositories, protective coating, combined LRW treatment, vitrification, plasma technology, cementation, state accounting system, radiation-ecological monitoring.

10.1 Introduction

Russia is one of only a few countries in the world to have all the elements of the nuclear fuel cycle, from uranium output to the complete set of facilities necessary for radioactive waste (RAW) management (Fig. 10.1). Russia produces 9% of the world's uranium output and 40% of the world's enriched uranium, supplying half of the uranium required for western design nuclear power plants (NPPs), and the Russian fuel company TVEL supplies 17% of the nuclear fuel used by NPPs for peaceful purposes. These NPPs produce 16% of the total electric power manufactured in Russia and form the joint stock company called the 'Concern Rosenergoatom'.

The global problem of nuclear and radiation safety is a historical inheritance from the Soviet atomic project. The accumulation of RAW and other nuclear materials since the Soviet era requires new approaches to the problem, including new methods both for processing and storing spent nuclear fuel and RAW and for decontaminating affected areas. Consequently, in 2007 the Russian government introduced a federal program

10.1 Map showing location of Russian nuclear facilities (courtesy of Rosatom state Nuclear Energy Corporation, Russia).

known as 'Nuclear and radiation safety assurance for 2008 and until 2015' with the specific aim of finding solutions to these problems. Plans are currently underway for the development of the atomic industry in Russia, which include addressing historical RAW issues and also taking into account the events of 2011 in Japan at Fukushima-1 NPP.

10.2 Special features of radioactive waste (RAW) accumulation in the USSR

The beginnings of the nuclear industry in the USSR can be traced back to the mid-1940s. The need for intensive and complex construction projects in the burgeoning atomic industry led to the introduction of a wide range of new techniques and procedures in different areas of science and technology within a short space of time, including: the search for uranium ore sources; the manufacture of enriched uranium fuel; the development of new construction materials, radiochemical technologies and measuring equipment; the creation of radiation safety systems; and the development and implementation of legal norms and regulations. These new and complex projects were carried out by a number of scientific and research institutes, special design bureaux, public health establishments, and by industrial enterprises and organizations across the former Soviet Union.

In the early stages of the development of the nuclear industry, scientific and research institutes were established in the major scientific centres of

the USSR, such as Moscow, Leningrad (now St Petersburg) and Gorky (now Nizhny Novgorod), with the aim of solving fundamental and applied problems related to the new atomic science. Important institutes such as the Kurchatov Institute (a National Research Centre), the Institute of Inorganic Materials, the Scientific and Research and Design Institute of Energy Techniques, the Institute of Chemical Technology, the Institute of Graphite Construction Materials, the Scientific and Research Institute of Physical Chemistry, and the Institute of Theoretical and Experimental Physics were all founded in Moscow and are still operational today. In 1923, V.I. Vernadsky founded the Radium Institute in St Petersburg, one of the oldest scientific and research institutes in the nuclear field, which investigates nuclear physics, radiochemistry, geochemistry and ecology, with specific reference to problems related to nuclear engineering, radio-ecology and the generation of isotopes. The institute actively participates in the implementation of federal programmes and international projects. Finally, the Scientific and Research Institute of Measuring Systems, founded by Y.E. Sedakov, is located in Nizhny Novgorod. As the number and complexity of studies grew, scientific and research institutes began to be moved into other parts of the USSR, including the Scientific and Research Institute of Technical Physics, founded by E.I. Zababakhin; and both the Scientific and Research Institute of Experimental Physics and the Institute of Physical-Energy, founded by A.I. Leypunskiy.

The production centres of the nuclear industries are also geographically widespread (sse Fig. 10.1). The principal Russian atomic production facility is the Mayak industrial association, which is the home of the Russian nuclear weapons programme, and the most extensive complex of interconnected production facilities, structurally divided into plants and production subdivisions, with reliable scientific and technical support. The complex also includes other important facilities such as the Mining and Chemical Industrial Unit, the International Center of Uranium Enrichment, the Machine Building Plant, and the Novosibirsk Plant of Chemical Concentrates. The infrastructure of many production facilities was based around what were known as 'closed cities' in Soviet times, and are now known as closed administrative-territorial formations (CATF), chosen for their location, layout and the make-up of their population. Today there are ten of these CATFs related to the Rosatom corporation.

The basic facilities of the nuclear industry that are subject to nuclear and radiation safety regulation in the Russian Federation, along with the number of each type of unit, are given in Table 10.1 [1].

In the early stages of the development of the new nuclear industry, a great deal of highly varied work was carried out simultaneously, from the design of RAW storage to immediate arrangements for the temporary placement of RAW. This simultaneous development meant that systems approaches

Table 10.1 Basic objects of nuclear and radiation safety regulation

Facility	Facilities and procedures subject to nuclear and radiation safety regulation	Quantity, units
Nuclear installations	NPP buildings, nuclear research units, nuclear powered ships (ice-breakers and lighter aboard ship carriers), industrial reactors, facilities for nuclear fuel production, complexes for scientific and research works with nuclear materials, facilities for processing nuclear materials (enrichment, chemical, metallurgical and hydrometallurgical production, etc.), complexes for spent nuclear fuel reprocessing, uranium mining complexes.	213
Storage locations for nuclear materials and radioactive waste (RAW)	Separate stores for spent nuclear fuel, stationary constructions for storage of nuclear materials, ranges for storage of dumped uranium hexafluoride, ship and industrial reactor installations, tank store for liquid high-activity wastes, reservoir storage facilities for liquid RAW, facilities for solid RAW storage, locations used for underground burial of liquid RAW, plants for liquid RAW processing.	454
National radiation sources	Medical, scientific and research laboratories and other facilities where open radionuclide sources are used, complexes, installations, apparatus, equipment and articles with sealed radionuclide sources (such as technological and medical irradiating units, flaw detectors, radioisotope equipment, radioisotope thermoelectric generators).	16,745
National locations used for storage of radioactive materials (RAM) and RAW	Interregional locations for the storage of radioactive materials (under the control of the organization 'Isotope'), other locations for the storage of radioactive materials storage located in national atomic energy facilities, interregional locations for the storage of RAW (under the control of the organization 'Radon'), RAW stores located in national atomic energy facilities containing only natural radionuclides.	1,508

for RAW management were impossible; moreover, there was a lack of overseas experience, and no defined strategy for successful practice. Questions of environmental security were not yet being prioritized, with the main requirement being the radiation safety of personnel dealing with RAW. Accidents leading to a release of radioactivity occurred at several

stages of the nuclear refining process. Procedures for eliminating the consequences of these events required the implementation of a temporary scheme for emergency waste management; temporary RAW storage facilities were therefore created using bulk design concepts.

10.2.1 The Eastern Urals Radioactive Track (EURT)

The danger of RAW as the cause of ecological disaster first became evident 12 years after the first use of nuclear weapons. Large-scale environmental pollution occurred due to the disturbance of RAW technologies at the Mayak nuclear facility in the USSR on 29 September 1957. An explosion occurred in a tank with a volume of 300 m^3 of which about 80 m^3 was high level waste (HLW). According to different estimates, up to 20 MCi of radioactive materials was ejected into the atmosphere [2]. The radioactive cloud, which consisted of liquid and solid aerosols, reached a height of 2 km. Radioactive substances fell over an area of more than 300 km, northeast of the point of impact (following the wind direction). The area of radioactive contamination was over 20,000 km^2, and included several of the facilities that made up the Mayak site. About 270,000 people in 217 contaminated areas of Chelyabinsk, Sverdlovsk and Tyumen' Oblast were subjected to different levels of radiation. This contaminated region is known now as the Eastern Urals Radioactive Track (EURT). The population of 23 villages was resettled away from the contaminated region, with over 12,000 people affected by the consequences of the explosion. To avoid the spread of radiation, in 1959 a government resolution determined the formation of a protective zone around the most contaminated part of the radioactive track, in which any economic activity was forbidden. In 1968 the Eastern Ural Radioactive State Reserve was established in the contaminated territory and still exists today.

Another example of radioactive contamination in this area is the discharge of liquid radioactive waste (LRAW) into the Techa river. In the summer of 1951, over 2.8 MCi of radioactive substances was discharged into the Techa [3]. The amount of radionuclide contamination entering the river later decreased, due to the curtailment of LRAW discharges and the building of weirs and bypass channels between 1951 and 1964. The comparative characteristics of the radiation exposure in the Techa river and in the EURT are given in Table 10.2 [4].

10.2.2 Formation of the institutional RAW management system in the USSR

Towards the end of the 1950s, along with the formation of the Eastern Urals Radioactive Track, the process of RAW accumulation began in the

Table 10.2 Characteristics of radiation exposure on the Techa river and EURT

Fundamental characteristics	Techa river	EURT
Summary discharge, Ci	3×10^6	2×10^7
Type of pollution	Aqueous	Air
Isotopic composition	^{90}Sr, ^{89}Sr – 20.4%, ^{137}Cs – 12.2%, ^{95}Zr+^{95}Nb – 13.6%, ^{106}Ru+^{106}Rh – 25.9%, REE – 26.9%	^{90}Sr+^{90}Y – 5.4%, ^{95}Zr+^{95}Nb – 24.9%, ^{144}Ce+^{144}Pr–66.0%, ^{137}Cs – 1.0%
Contamination area	The Techa and the Iset rivers(width to 4 km)	23,000 km^2 (density 0.1 Ci/km^2)
Maximum density of contamination	10,000 Ci/km^2 (by ^{137}Cs)	3,000 Ci/km^2 (by ^{90}Sr)
Maximum exposure dose rate	3.5–5 Rh/h	1–3 Rh/h
Taken out of land tenure	80 km^2	1,000 km^2
Number of people exposed to radiation	12,400 people (Techa and Iset rivers)	272,000 people
Displaced population	about 8,000 people	More than 12,000 people

territories where scientific and research institutes were located, mainly in the large cities in the central part of the USSR. Furthermore, the development of the nuclear industry helped to solve a wide range of medical and technological problems with the help of radioactive materials. Two new systems were almost simultaneously implemented by government resolution.

The first of these systems was 'Isotope', an All Union association established in 1958 with the aim of revolutionizing isotopic production, radiation techniques, and medical and general purpose equipment.

Isotope had the following functions:

- delivery of isotopic products for external and domestic markets;
- addressing radiation safety problems during handling of ionizing sources (IS);
- supplying medical and scientific establishments with the required isotopic products;
- development of radioisotopic technologies.

The establishment of this organization helped to solve many of the problems associated with the introduction of new nuclear technologies and their influence on the national economy.

At the same time, a centralized system for the collection and disposal of RAW and spent ionizing sources (SIS) was created, with 35 different

organizations involved (16 from the Russian Federation, 5 from Ukraine, and 1 from each member republic of the USSR). For example, in February 1960 the Council of Ministers of the USSR created an organization called 'Radon' in Moscow, which was designed to act as a central facility for RAW processing and disposal serving organizations in Moscow itself, the Moscow region and 10 adjacent regions. It began practical operation at the start of 1961, when the special vehicles column made its first journey to the Kurchatov Institute.

The introduction of these new specialized facilities for RAW and SIS collection and disposal stabilized the accumulations of RAW in scientific and production establishments across the USSR, as RAW removal began to be effectively and routinely carried out. For example, industrial, medical and research establishments in the central regions of the USSR sent the following quantities for further disposal:

- up to 2,500 m^3 of solid radioactive waste (SRAW) with an activity up to 10^{15} Bq,
- up to 300 m^3 of liquid radioactive waste (LRAW) with an activity up to 10^{11} Bq,
- up to 20,000 units of SIS with an activity up to 10^{16} Bq.

The sources were predominantly composed of ^{60}Co (more than 90% of the overall activity) and ^{137}Cs (up to 6%). Over almost 50 years, more than 100,000 m^3 of RAW was removed from the Moscow area. These specialized enterprises also improved radiation control systems, developed monolithic matrix technologies as a product of RAW processing, and drew up new models and algorithms for safe RAW processing.

10.2.3 Establishment of the Moscow Science and Production Association (MosNPO) 'Radon'

In 1971 the central radiation safety organization, Radon, looked at the total of its results over the first ten years of operation. The enterprise already had a well-developed infrastructure, skilled staff, methods and technologies for RAW localization and a substantial amount of operational experience. Hot cells equipped with manipulators were put into operation and were allowed to operate remotely in RAW and SIS procedures. In the same year, the director of Radon (I.A. Sobolev) prepared a doctoral thesis dedicated to the problems of RAW cementation, which he successfully presented to the scientific and technical council of the Institute of Inorganic Materials under the auspices of A.A. Bochvar. To allow the routes used by the special RAW transport to be monitored for radiation levels, two mobile radiometric laboratories were set up. The activity of Radon then became known abroad: the first technical visits by foreign representatives of the atomic

industrial forum of Japan, parliamentary groups from Sweden and scientists from Canada took place in 1975.

From 1976 to 1980 new facilities were installed at Radon for RAW bituminization (URB-200), combustion (USGO-80) and compaction (BA-1330). During the same period, a facility for water purification (EDU-500) was assembled, and test runs were carried out. Another considerable achievement was the implementation of a pilot industrial facility for RAW combustion or thermal treatment: RAW combustion was carried out for the first time in the history of Soviet industrial tests in NPPs. The results of these tests formed the basis for the design and creation of the technological complex for the Kursk NPP. The application of a 'dry' system for the treatment of off-gases helped to solve the problem of secondary LRAW generation. To obtain bitumen compounds, the UBD-200 facility was developed, with elements of mechanization and automation for the whole process from RAW loading to the unloading of the end product (i.e., the bitumen compound).

Radon had swiftly become a multi-purpose scientific and technological complex. The new technological developments introduced at Radon were used at other facilities for RAW processing and disposal. In 1977 the State Committee on Inventions and Discoveries awarded two certificates of authorship to the staff of Radon for patented inventions. Radon's main achievement in 1978 was the establishment of the new high-capacity facility for LRAW bituminization (URB-8). Work was also carried out on the selection of the correct composition for LRAW vitrification. Under the management of the corresponding member of the Russian Academy of Sciences and the director of the Institute of Inorganic Materials (A.S. Nikiforov), the process of high-level LRAW vitrification at hot cells was investigated, using simulators of high level LRAW from Mayak. In 1979 the containment facility for HLW and SIS was completed. Research into hot cells was carried out, including studies into the content of intermediate and high level waste. Experiments into waste vitrification were continued, alongside new studies into plasma methods, and a new facility for RAW combustion, known as Fakel, was completed.

In 1980, the Council of Ministers of the USSR converted the enterprise into the Moscow Science and Production Association 'Radon' (MosNPO Radon); after that, the organization became responsible for the methodical and scientific management of RAW at special nuclear facilities across the USSR. In 1984, before the Chernobyl disaster, the government of the USSR charged MosNPO Radon with the organization and implementation of detailed radiometric monitoring of Moscow city and the Moscow region in order to define areas with possible radioactive contamination. Furthermore, in 1986, MosNPO Radon, Minsredmash and the USSR Academy of Sciences were jointly entrusted with scientific research and experimental

design with the aim of improving RAW processing technologies such as combustion, vitrification, cementation, etc.

10.3 Long-term RAW isolation strategy

The RAW isolation process is based on the principle of multi-barrier protection at all stages. RAW conditioning allows the immobilization of radionuclides in durable matrix materials (such as glass, cement, ceramics, etc.), which are then placed into special protective casks, such as metal drums or metal and reinforced concrete containers, for transportation. Any voids are filled with special backfill. Conditioned RAW is then placed into hydroisolating repositories, of which there are three types: near surface, well-type and drill-type. These constructions are supplied with a system of multifunctional barriers, which prevents any interaction between RAW and external factors. Interaction between the radionuclides and the environment is prevented by both man-made and natural barriers, with each performing its own shielding function. The multi-barrier system means that the safety of the repository is not dependent on one barrier alone, and is assured not only by technical means, but also by technical-organizational measures. The principle of multi-barrier protection guarantees the safe storage of RAW over the whole period during which they pose a radiological hazard.

In accordance with the recommendations of the International Atomic Energy Agency (IAEA), long-term RAW storage facilities must guarantee geo-ecological safety for the entire period of operation. In the national programmes of RAW management, most countries have set the normative period for low and average level activity RAW at 300 and 500 years. This can be explained by the fact that near-surface type repositories are only used for low- and intermediate-level RAW (LLW and ILW) with half-lives of less than 30 years. Over the course of the operation of these repositories, the activity due to natural decay will be lowered, in comparison with the initial level, by three orders for LLW and five orders for ILW.

10.3.1 Construction of 'historical' RAW repositories

As mentioned above, facilities for RAW storage (of the Radon type) are intended for the long-term storage of RAW containing short-lived radionuclides with a half-life of less than 30 years, including ^{137}Cs and ^{90}Sr. They only contain LLW and ILW. The RAW suppliers are the nuclear facilities of the nuclear industry (unconnected to the fuel cycle), organizations that operate nuclear reactors for research, and medical, training and scientific research centres that carry out radioisotope production.

The selection of suitable sites for RAW repositories was conducted firstly on the basis that any transfer of radionuclides into underground flowing

water or its environs must be avoided. RAW repositories must therefore be placed in a clay massif, with low filtration and high sorption properties for radionuclides. The distance to the nearest water-bearing horizon must exceed 10 m. An area that met these requirements was identified 25 km from the town of Sergiev Posad in the Moscow region. From a geological point of view, the area offers sturdy layers of clay deposits of glacial origin (the Moscow and Dneprovsk moraines) that limit filtration and provide high sorption. The nearest water-bearing horizon within the limits of this area is located where the deposits of the Moscow and Dneprovsk glacial moraines meet at depth intervals of 38–42 m [5]. Groundwater in the area is not subject to regional propagation and remains local to the site, appearing only during autumn and winter in the areas adjacent to the man-made constructions. The filtration factors of the glacial clay deposits vary between 0.001 and 0.003 m/day, depending on the presence of sandy interlayers and the disturbance of the integrity of the base soil; these soils also have a high sorption capacity. Given that more than 90% of RAW contains ^{137}Cs with a half-life of less than 30 years, ^{90}Sr (29 years) and ^{60}Co (5 years), the composition of the soil makes the selected area an ideal location for RAW repositories, meaning that the radiation safety of the population outside the limits of the facility's protection zone was guaranteed.

The construction of near-surface repositories for LLW and ILW began in the mid-1960s. A number of advances in repository design have since taken place, which have improved hermetic conditions and allowed the creation of reliable monitoring systems. Some key developments are:

- sunken monolithic repositories with a capacity of 400 m^3, a standard 1960s project designed by the USSR State Special Design Institute of Minsredmash (SSDI), a historical repository;
- sunken composite repositories, a historical type of repository designed by MosNPO 'Radon';
- sunken composite repositories with a ground-based tier, a 1980s development by MosNPO Radon;
- sunken monolithic repositories with a capacity of 5,000–10,000 m^3 (SSDI);
- sunken monolithic repositories with a hangar superstructure, developed in the 1990s (SSDI);
- large diameter boreholes;
- drill-type repositories for SIS of the SSDI;
- repositories for SIS containing ^{226}Ra, developed by MosNPO Radon.

Near-surface type historical repositories take the form of trench grooves 4–5 m deep, with the bottom of the trench covered with a hydro-insulating layer. The walls are made of reinforced concrete blocks or monolithic reinforced concrete 0.4 m thick. The top of the repository is covered with

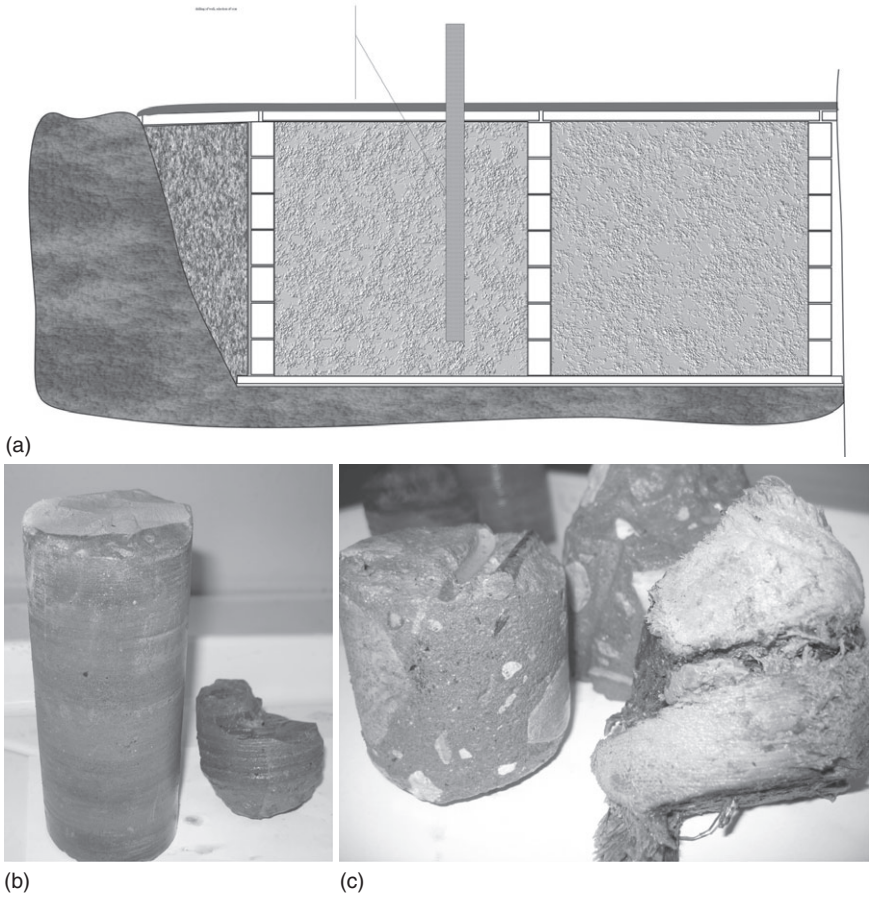
reinforced concrete slabs and a layer of asphalt. Internally, the repository is made up of several sections. RAW in the repository was bulked in 1 m thick layers, which were then plugged with cement solution, prepared for use with LRAW with low salinity. In the mid-1980s an additional level, 3.5–4 m high, was built above some of the repositories, creating two-storey constructions with a capacity of about 20,000 m^3.

Up until the 1990s these repositories were considered disposal facilities and were intended for the final disposal of LLW and ILW, with the surrounding rocks carrying out a basic barrier role to ensure geo-ecological safety. From the end of the 1990s, following IAEA recommendations, these near-surface type constructions were given a new status as 'RAW repositories with a limited period of storage'. The new designation was based on the idea that adequate environmental protection from the hazards of RAW is dependent on man-made barriers, i.e. durable matrixes, RAW packing, backfill between packages, and the structural elements of the repository.

10.3.2 Restoration of the historical repositories

The main advantage offered by RAW storage is the guaranteed security afforded by the integrity of each separate element of the multi-barrier complex. If any one of these multiple shielding barriers is damaged, the potential risk of migration of radionuclides into the environment increases. Any number of factors can disrupt the integrity of the construction material of the repository and/or of the massif of solidified RAW, principally: (1) the deformation of structural elements as a result of significant temperature variations in the environment (e.g., seasonal freezing and thawing) or as a result of shrinkage; (2) the decompression of seams; and (3) any construction defects including where the different elements meet, such as joints between sides of walls and partitions and the upper overlap. In addition, cracks and microcracks can appear on the surface of the construction (the outer duct), which may be connected to each other, leading to the formation of a system of interconnected channels in the walls of the repositories.

An estimation of the conditions of the man-made barriers, carried out in the early 2000s [6, 7], included comprehensive geological, hydrogeological, geophysical and radiometric studies of the condition of the RAW massif, the soils forming the edge zone and the massif of the surrounding rocks. In the selected zones boreholes 8 m in depth were drilled, and various measurements were taken: the layer-by-layer permeability of the medium was measured; gamma- and thermo-logging were carried out; and the core material (RAW cement compound and soils, as shown in Fig. 10.2) and water was sampled for further radiochemical, chemical, physical, mechanical, micro-structural and microbiological analysis.



10.2 (a–c) Sampling of cemented RAW from the near-surface repositories.

These comprehensive studies, carried out with the help of boreholes drilled in the near-contour zone, determined that the high sorption property loams of the near-contour zone are indeed a reliable obstacle that prohibits the radionuclides from entering the adjacent massif [8]. During the investigation of the cement compound samples containing RAW (core material), it was established that a cement matrix that has been age-hardened for more than 40 years generally preserves its basic immobilization properties. The majority of samples tested (Table 10.3) have a compressive strength higher than the normative value of 5 MPa [9] and the rate of radionuclide ^{137}Cs leaching is lower than the required level of 1×10^{-3} g/(m^2·day). However, sufficient humidity and porosity together with structure friability and low compressive strength attested in several samples from different depths were

Table 10.3 The physico-chemical properties of the samples of the cement compounds with RAW from near-surface repositories

Sample description		Specific activity $\Sigma\beta$ on ^{137}Cs (Bq/kg)	Porosity (%)	Humidity (%)	Compressive strength (MPa)	Leaching rate ^{137}Cs (g/(m^2·day))
Dense cement block with cellulose inclusion, without visible damage (1960, encapsulation in 2004)		3.46×10^7	19.5	5.6–8.9	7–18	5.3×10^{-4}
Scattering or friable cement block with wood and rubber inclusions and cracks, crumbles (1961), at depth, m	0.1–1.0	$(1.2–6.5) \times 10^7$	20.1	25.9–38.1	5.7–23.9	$(1.61–2.4) \times 10^{-3}$
	1.0–2.5		44.2		3.9–13.0	$(1.98–3.2) \times 10^{-3}$
	2.5–3.8		36.4		2.3–10.2	$(0.66–7.4) \times 10^{-4}$
	3.8–6.5		28.0		1.3–3.5	$(0.58–7.9) \times 10^{-5}$
	6.5–8.0		11		0.3–1.4	$(1.06–5.2) \times 10^{-2}$
Without visible structure damage (1965)		$(0.16–7.85) \times 10^6$	32.8	15.9–32.5	14–20	$(0.28–9.4) \times 10^{-4}$
Without visible structure damage (1987)		1.59×10^6	17.7–32.3	2.30–30.0	5.3–5.6	1.6×10^{-6}

also observed, showing that destructive processes were taking place in the cement matrix [10].

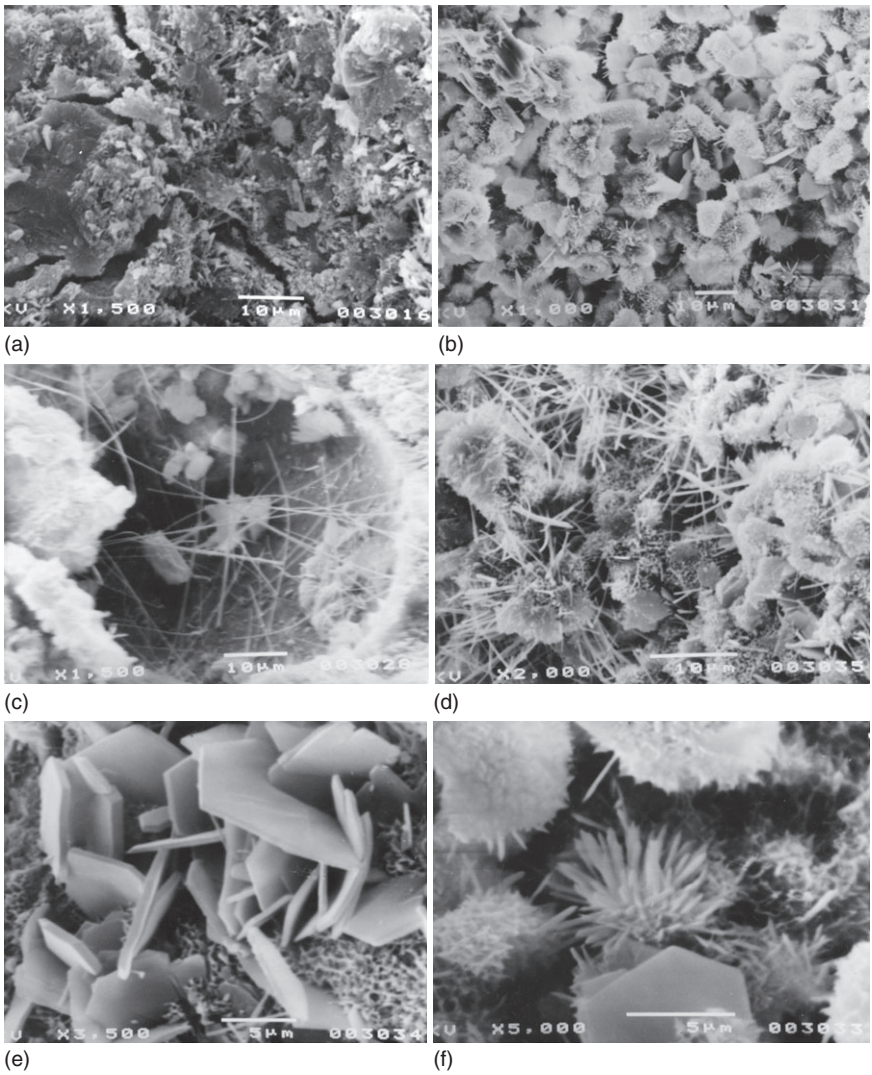
Microbiological investigations found that the cement compounds, the pore water in the cement compound and the penetrating waters contained microorganisms of different physiological groups, such as anaerobic fermenting, denitrifying, nitrifying or sulphate-reducing bacteria, as well as fungi, which were capable of destroying silicate materials (Table 10.4). Studies were carried out to determine the form and number of bacteria and produced metabolites as well as the influence of these metabolites on the microstructure and immobilization properties of the cement matrix [9, 11].

RAW intended for cementation contains components that may provide nutrition for microorganisms, which can then grow under the favourable conditions offered by near-surface repositories, such as insufficient exchange of air, humidity, temperatures of 6–28°C, and a pH of between 5 and 9. The main nutrients for microorganisms are provided by LRAW in the form of aqueous solutions of nitrates, sulfates and chlorides at concentrations of up to 300 g/l, mineral oils and organic liquids (extractants, scintillators), and also wood, plastic and cloth remnants. Gas-liquid chromatography was used to determine that the denitrifying and fermenting bacteria are capable of secreting $(8.6–10.6) \times 10^{-2}$ ml of N_2 within 24 hours from 1 cm^2 of the RAW cement compound surface.

The products of the metabolism of microorganisms can cause changes in the way in which cement hydrated minerals crystallize, as well as disturbances in the cement matrix microstructure. Consequently, the microstructure of samples of cement compound cores taken from more than 4 m below the surface was examined. At this depth, the action of aggressive factors such as freezing and the penetration of surface and groundwater are not factors. The cores were examined using petrography and scanning electron microscopy (Fig. 10.3). Numerous local sections of the cement matrix displayed internal cracks (Fig. 10.3a), and 20–200 μm micropores in which thin long fibres of new mineral formations were nucleated (Fig. 10.3c). The resultant body of hydrated cement minerals was not the uniform dense structure expected for this significant period of hydration, but a friable 'corroded' structure (Fig. 10.3b, d). The products of the hydration of calcium silicates, which determine the strength of the cement matrix, are heterogeneous in their composition (refractive index N = 1,573–1,590). Calcium aluminate hydrates are heterogeneous in size (2–50 μm), represented by heterogeneous cubic and hexagonal crystals (Fig. 10.3e), surrounded by single ferrite gel. This is the characteristic crystallization nucleation of needle-shaped and rhombic crystals of ettringite $3CaSO \cdot Al_2O_3 \cdot 3CaSO_4 \cdot 32H_2O$ and calcite $CaCO_3$ (Fig. 10.3d,f), which fill the cement pores and bridge the crack that has been generated [12]. Destruction of the type observed here

Table 10.4 Microbiological characteristics of the samples of cement compounds with RAW from near-surface repositories (conservation in 1960–1961, 1965, 1987)

Physiological groups of microorganisms and their characteristics		Number of bacteria, isolated at		
		Samples of cement compound, kl/g	Samples of soil, kl/g	Samples of ground water, kl/ml
Total amount of microorganisms (colony forming units/g or ml)		9.1×10^2 … 1.9×10^4 CFU/g	4.9×10^6 CFU/g	1.4×10^5 CFU/ml
Nitrifying bacteria		0	1.5×10^2	0
Denitrifying bacteria	1 phase	0	1.2×10^5	1.3×10^4
	2 phase		3.6×10^4	6.0×10^4
Sulphate-reducing bacteria		2	1	0
Fermenting bacteria		60 … 3.0×10^4	3.6×10^5	2.5×10^2
Iron-reducing bacteria		0	2	2
Iron-oxidizing bacteria		33	0	0
Thione bacteria		0	0	0
Micromycetes		45 … 1.5×10^4	2.6×10^6	8.0×10^4 CFU/ml
Generic assignment		*Bacillus Pseudomonas Rhodococcus, Alcaligenes Micrococcus Mycobacterium Arthrobacter*	*Bacillus Pseudomonas Rhodococcus*	*Arthrobacter Rhodococcus Alcaligenes Rhodococcus Bacillus Flavobacterium*
Properties of strains	Microscopic picture	Rod-shaped bacteria, in pairs or singles, containing spores	Rods, cocci	
	Grams-stain	Gram +	Gram +	Gram –, Gram +
	The reaction with catalase	+	+	+



10.3 Internal chippings of cement compound core from near-surface repositories of 1960–1961 (microphotography by scanning electron microscope): (a) internal cracks; (b) friable structure, (c) pores, filled with the fibrous new formations, (d) needles of ettringite, (e) hexagonal plates of hydro-aluminate, (f) the mutual germination of the needle-shaped and rhombic crystals of ettringite and calcite.

may be the result of neutralization and carbonization caused by the biogenic acid products of the alkaline minerals in the cement matrix, mainly portlandite $Ca(OH)_2$ and the hydrosilicates of calcium, and of recrystallization and an anomalous growth of ettringite crystals in the space of the microcracks of biogenic origin [13].

A number of biocidal additives are being investigated as possible means of preventing microbiological destruction in cement compounds. The minimum concentration of these additives required to suppress the growth of the bacteria characteristic of near-surface repositories is 0.0015–0.003 mg/ml, and for complete disinfection, 0.003–0.006 mg/ml. Effective biocides of the poly-hexamethyleneguanidine class are chemically compatible with the components of the cement solution [14, 15], allowing the development of the highly penetrating cement compositions known as 'Bison – BPl' and 'SPCK', which were successfully tested in 2003–2007 during the hermetic sealing of RAW near-surface repositories from the 1960s. In the RAW technological cementation processes, these compositions [16, 17] were first used in 2004 to prevent undesirable microbiological processes in repositories for solidified RAW.

To correct the disturbances formed in the man-made barriers of the historical near-surface repositories, a method of repeated grouting was developed for cemented RAW massifs. The method relies on boring into the RAW cement massif forcing a mixture of biocidal and highly-penetrating cement compositions such as Bison – BPl and SPCK into the cement mortar. This repeated grouting allows the restoration of the impermeability of the repository and prevents the migration of radionuclides out of the construction. An upper layer of protective coating was also developed [6] with the aim of counteracting the influence of sediments and seasonal temperature differences on the construction of near-surface type repositories and their contents [6]. This coating is a multilayer screen 2.7–3 m thick, made mainly from natural materials, that cover the entire surface of the construction. The stages in the creation of this sealing coating are presented in Fig. 10.4.

The creation of protective coating for historical RAW repositories involves the following steps:

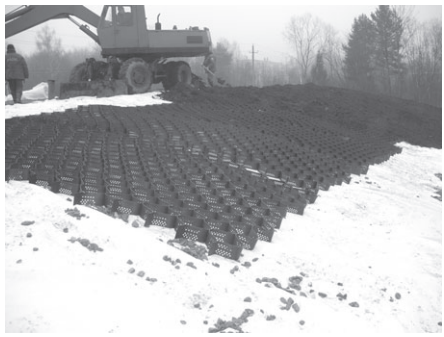
- repeated grouting using highly penetrating biocide cement compositions of cemented solidified RAW;
- creation of an anti-filtration screen in the soil on the perimeter of the repository;
- formation of a barrow from the natural materials on the surface of the repository;
- piling and welding sheets of the geo-membrane Carbofol (1.0–2.0 mm thick);

(a)

(b)

(c)

(d)



(e)

(f)

10.4 (a–f) Complex of works of preservation coating for 'historical' near-surface RAW repositories.

- creation of drainage systems for rainfall outlet;
- creation of a system of boreholes for observation and radio-ecological monitoring of the repository;
- creation of drainage and protective layers of a preservation coating from sand, clay, gravel and topsoil.

10.4 A new type of repository: the adoption of the controlled prolonged storage concept

In 2010–2011 the construction of a new type of RAW repository was completed. This new repository is a ground-based reinforced concrete bunker with an area of $172 \times 190\,m^2$ and a height of 6 m, divided into 20 autonomous modules. In this construction, the boundary area between the RAW massif and the natural geological medium is reduced, the repository is reliably protected from rainfall and flooding, maintenance and monitoring is simpler, and RAW can be localized with the possibility of retrieval.

Each bunker is designed to store $110{,}000\,m^3$ of conditioned RAW, immobilized into a cement matrix, packed into long-life reinforced containers (NZK-150-1.5P), and sealed using a special non-shrinking concrete mixture. Containers are brought into the bunker on trucks and are placed into the relevant sections using a forklift truck. The repository has a ventilation system in order to remove off-gases, along with three drainage systems (rain canalization, near-wall-bed drainage, and a system for filtrate removal). RAW can be safely stored for 50 years in this ground-based bunker, with RAW that may need to be moved to another location after 50 years stored separately.

If the facility is to become a disposal site, the free space between the packages containing conditioned RAW is filled with bentonite, which is able to absorb radionuclides and swells on contact with water, thus filling any cracks and repairing other possible damage caused by construction sagging. The construction is covered by an elevated multilayer protective coating, which includes a biological barrier and a watertight screen consisting of reinforced concrete, and layers of crushed stone, clay, geo-textile, geo-membrane, silt, sand and topsoil. The design of this construction guarantees the long-term safe storage of conditioned RAW.

10.5 Survey of modern RAW management technologies

RAW management requires a systemic approach: all the stages of RAW management are the components of one overall system, and all the

techniques and procedures adopted in each separate stage are connected and have a common goal of radiation safety.

10.5.1 RAW management system

Russia adheres to the principles of safe RAW management [18], involving the following basic stages of the process:

1. Pretreatment
   - collection
   - segregation
   - chemical adjustment
   - decontamination.
2. Treatment
   - volume reduction
   - removal of radionuclides from the waste
   - change of composition.
3. Conditioning
   - solidification
   - immobilization
   - overpacking.

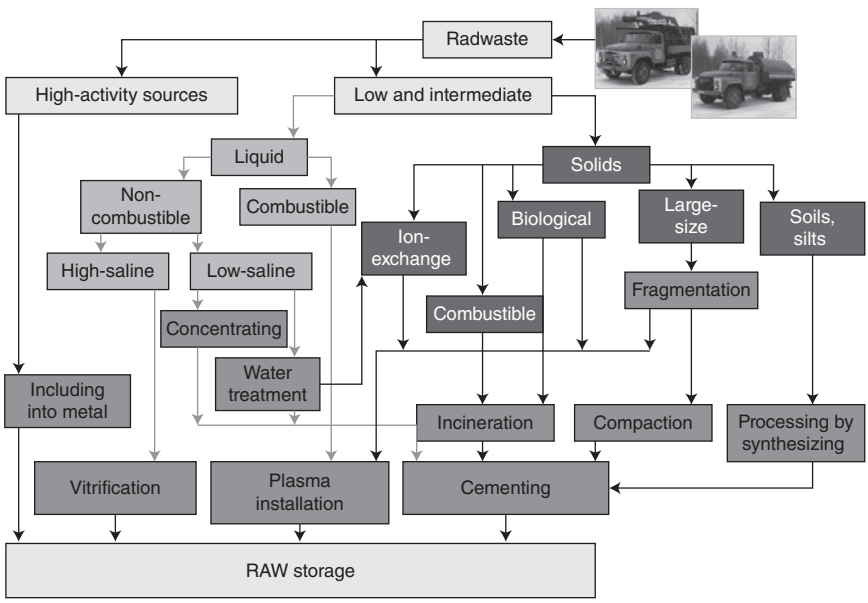
RAW is divided into categories based on its origin; its physical, chemical and biological properties; its state of aggregation; and level of activity. The different properties of different types of RAW make it necessary to use different RAW management technologies for different sorts of waste. In Russia, a number of technological methods have been developed to allow the optimal processing of RAW. The basic methods used for RAW processing [19] are presented in Table 10.5 and Fig. 10.5.

LRAW constitutes the majority of all RAW in Russia, with 90% of LRAW made up of aqueous solutions originating from (a) technological drains, which are produced at industrial and research centres, including medical and biological laboratories; and (b) decontamination drains from the decontamination of equipment and overalls. These aqueous solutions are highly varied in their chemical and radionuclide composition, and include such diverse forms as ions, dissolved complexes of organic substances, colloidal particles and micelles, suspended solids, and liquid emulsified oil products. Solid RAW includes different materials contaminated with radionuclides such as construction materials, dismantled equipment, spent filters and resin, corpses of experimental animals, and silts and soils from contaminated territories.

The basic aim of RAW treatment is to reduce the physical volume of the waste and to transfer this waste into a monolithic, chemically and mechanically stable form, suitable for long-term storage in containers.

Table 10.5 The basic methods for RAW processing

RAW Type		Processing methods
Solid	Combustible	Combustion in furnaces on fire grates at 900°C, plasma treatment, thermochemical treatment, vitrification, acid decomposition
	Compactable	Compaction at low and high pressure, super-compaction
	Metallic	Compaction, melting
	Incombustible and non-compactable	Direct placement into containers
Liquid	Organic combustible	Combustion, joint combustion with SRAW, encapsulation in cement matrix
	Organic incombustible	Absorption using powders and encapsulation in cement matrix, thermochemical treatment
	Liquid low salinity	Purifying (concentration) by evaporation, by chemical precipitation, by absorption, by selective absorption, by a membrane separation process, by cementation
	Liquid high salinity	Purifying by selective absorption, cementation, bituminization, vitrification
Gaseous		Trapping by absorption and through the use of chemical reagents



10.5 Basic methods of RAW treatment at MosNPO 'Radon' [19].

10.5.2 Combined LRAW treatment

The optimum treatment of low-level liquid RAW involves a modular approach to the process, in which there is an autonomous technological module for each specific technology used in the LRAW treatment. At MosNPO 'Radon', long-term investigations were carried out into the effectiveness of different methods of low-level liquid RAW treatment, resulting in the development of a modular unit known as 'Aqua-express', involving h sorption and diaphragm methods of cleaning [20]. This unit, shown in Fig. 10.6, includes a filter-container with a nickel ferrocyanide absorbent (1), a cascade of sorption-filled filters (2), an ultra-filtration module (3) with membranes, the pore size of which does not exceed 50–100 nm. The unit's capacity is 300–500 l/h of initial liquid RAW.

In 2002–2006 modular units of this sort were supplied to research centres in Bangladesh, Syria, Iran, Serbia and Uzbekistan. In 2006–2007 modular units were included in the projects of the Kazan department of the Federal State Unitary Enterprise (FSUE) 'RosRAO' and the United Institute of Energy and Nuclear Research in Belarus, and in 2007 they were delivered to Rostov SK Radon.

10.5.3 Vitrification of LRAW

In Russia, LRAW vitrification on an industrial scale is performed only at Mayak and MosNPO 'Radon'. At Mayak, this is achieved by direct (or 'Joule') electric heating in crucibles with submerged electrodes (pot furnaces with refractory linings), while at 'Radon' induction heating is used,

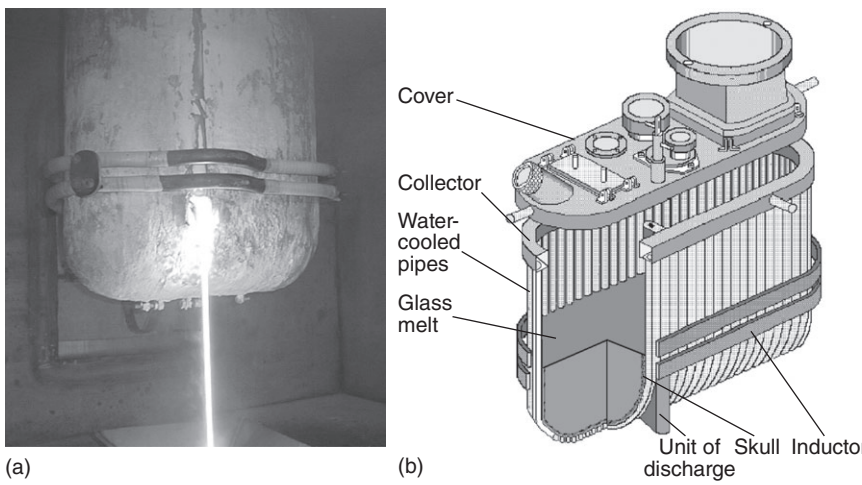
10.6 Mobile modular installation 'Aqua-Express' for LRAW treatment.

whereby the crucible is surrounded by water-cooled metal tubes (known as the 'cold crucible' method).

Vitrification of high-level LRAW at unit EP-500 of the Mayak plant was started in 1991 and relies on the vitrification of RAW into a sodium aluminophosphate glass by means of direct electric heating [21]. The ceramic melter in EP-500 has a rectangular configuration with vertical molybdenum electrodes at the bottom lined with refractory blocks (predominantly ZrO_2 and Al_2O_3). The operating temperature is 850–900°C. Finished glass is periodically poured into 200 L canisters, three of which are loaded into one container and are placed into the controlled ground-based storage. Approximately 23,000 m^3 of LRAW with an overall activity of 495 million Ci has been processed since 1987.

The main advantages offered by ceramic melters are technological flexibility, which makes it possible to include LRAW in glass with highly varied compositions, and the uniform heating of the glass melt. The result is a high quality and homogeneous end product. Furthermore, during the vitrification process the ceramic melters cause less volatilization of radionuclides due to charge zonal heating. On the other hand, the disadvantages of ceramic melters are the complexity of their construction, the low corrosion resistance of the materials, the large overall size and mass, the need for continuous operation, and the complexity of repair, replacement and decommissioning.

At MosNPO 'Radon' the vitrification of liquid RAW [19] is conducted with a 1000 kW cold crucible melter with a capacity of up to 0.3 m^3/h, and a final glass production rate of up to 75 kg/h (Fig. 10.7). The melter is placed



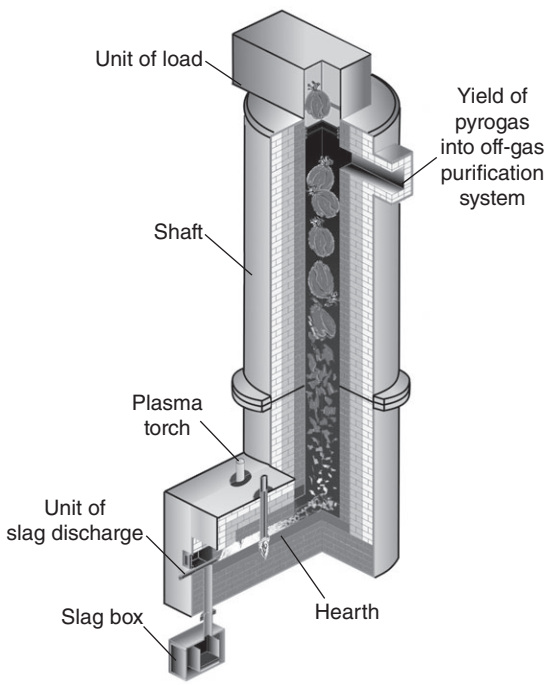
10.7 (a, b) Cold crucible for installation of liquid RAW vitrification.

inside the copper inductor using induction heating, together with the capacitors of the oscillatory circuit, which are connected to the generator. The absorption of the electromagnetic radiation created by the generator, takes place in the material inside the crucible and its walls, which are cooled by water.

10.5.4 RAW plasma treatment

The newest method of high-temperature SRAW treatment is plasma technology using a plasma pyrolysis treatment unit (Fig. 10.8.)

At MosNPO 'Radon', an industrial plasma treatment unit for mixed RAW treatment has been put into operation, with liquid slag disposal and a capacity of up to 250 kg/h [22, 23]. This facility makes it possible to obtain a durable glassy end product in only one step, which is suitable for long-term storage or disposal without further treatment. Plasma treatment, in contrast to combustion technology, can treat not only combustible waste, but also incombustible components such as construction materials, glass, soil, metal scrap and thermal insulation materials. The specific activity of



10.8 Installation based on use of high-temperature furnace with plasma heating.

the RAW is limited to the values of 3.7×10^6 Bq/kg for β- and γ-emitting radionuclides and 2.2×10^5 Bq/kg for α-radiating isotopes.

Plasma treatment currently allows the treatment of morphologically different types of RAW at the expense of RAW with high humidity, such as soils, ion-exchange resins and spent ion-selective absorbents. The overall RAW humidity was within the limits from 15 to 40%, but the humidity of RAW in any one package can reach 90%. The proportion of incombustible components in one package can be as much as 40%.

The plasma treatment unit called 'Pluton' has a shaft furnace, which consists of a shaft and a melter, and units for RAW loading and molten slag discharge. There are two 100–150 kW plasma torches in the arch of melting camera, which maintain the melting temperature in the 1500–1700°C range. The temperature of the off-gases does not exceed 250–300°C, which is reached due to the heat absorption of overheated gases during waste drying and pyrolysing. At the same time, the retention of volatile radioactive and chemically toxic substances (heavy metals) occurs in the middle and upper waste layers in the shaft furnace.

The reduction factor of the initial volume of RAW with respect to the volume of slag obtained varies from 66 to 120 and depends on the morphology of the waste. The receptacles containing the slag compound are placed into shielded irretrievable containers which are then placed into long-term storage. The reduction factor in waste volume, taking into account the placement of the final conditioned product into containers, is from 40 to 70 [24].

10.5.5 RAW cementation

The basic industrial method used for the conditioning of low- and intermediate LRAW is cementation, which offers a durable and incombustible end product, requires little capital investment and operational expenditures, and satisfies the quality requirements of the Russian Standard. At MosNPO 'Radon', solid RAW is conditioned using a method relying on the impregnation of friable fine-grained or granular materials by a highly penetrating cement solution [25]. Cementation by impregnation allows the treatment of non-compactable, incombustible fragmented SRAW, finely dispersed ash residues from the RAW combustion process (with a predominant particle size of 0.05–10 mm), as well as tightly packed, fine-grained or mixed (in size and form) RAW.

As a rule, the void content in friable solid RAW makes up 40–45% of the bulk volume. Filling the voids between the particles with cement solutions forms a cement matrix, allowing an end product to be obtained with a volume equal to the initial bulk volume of the wastes. This in turn allows up to a twofold reduction in the required volume of the repositories used for cemented RAW, the simplification of the cementation process itself, a

reduction in cost, and an increase in the radiation safety of the technological process.

The method essentially consists of impregnating SRAW bulk volume with cement solution inside a container. A perforated probe is introduced into the lower part of the container, through which cement solution is delivered at low pressure (<0.1 MPa). As the cement solution rises evenly from bottom to top, it displaces the air, fills all voids and pores and then hardens, forming a monolithic cement compound.

This impregnation method offers a number of advantages over traditional cementing:

- an increase in the degree of filling of the cement compound with solid withdrawals, which comprise ~ 50–60% of the total mass;
- the volume of the end product does not increase compared with the initial bulk volume of SRAW;
- the stages of sorting, dosing, crushing and mixing of solid radioactive wastes with cement mortar are not required as part of the process;
- the process does not require complicated expensive equipment;
- the volume of secondary liquid waste is lower than that from traditional cementation.

Table 10.6 provides some data on the influence of the different components of high-penetration cement solution (HPCS) on the parameters of impregnation and the end product strength.

An experimental industrial facility for the cementation of finely dispersed ash residues (resulting from RAW combustion) by impregnation in containers (barrel: 100l, 200l) has been developed at MosNPO 'Radon' [17]. The cementation unit and the end product [26] are presented in Fig. 10.9. The end product is a uniform cement compound, chemically and physically durable, whose quality satisfies all the requirements of the Russian State Standard GOST R 51883–2002. When complete, it is sent for storage in the initial container, which provides an additional shielding barrier.

Cement compounds obtained by the impregnation of ash residue from the RAW combustion unit have the following characteristics:

- they contain 55–70% RAW by mass;
- they have a compressive strength from 6 to 18 MPa;
- they have a leaching rate of ^{137}Cs in the range of 10^{-4}–10^{-3} g/(cm^2/day); and
- they are stable when exposed to temperature variations and water corrosion.

In addition to the impregnation of ash residues from RAW combustion, which are placed in 200 L barrels, other solid RAW can also be cemented

Table 10.6 Properties of high-penetration cement solution and final cement compounds with RAW (ash residue) obtained by impregnation

Composition of cement material in HPCS	Penetrating power of cement mortar		Strength of final cement compound with ash residue (63% by mass) at the top of 200 l barrel (after 28 days of hardening)*, MPa
	W/C in the initial cement solution	W/C in the cement solution after impregnation of ash (V = 200 l)	
Cement solution based on traditional Portland cement	0.8	Impregnation is impossible because of the choking of channels with cement particles	
	1.4	2.20	up to 1
70% Portland cement+29% fine-ground cement+1% PGMG polymer**	0.6	0.68	7–10
99% fine-ground cement+1% PGMG polymer	0.9	0.91	8–12
	1.1	1.20	6–8

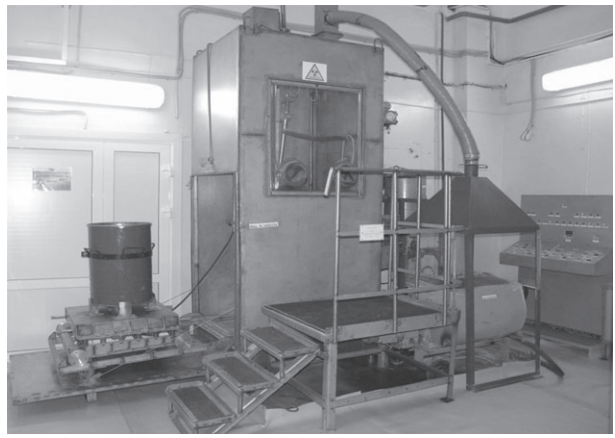
* Values vary because of the assorted composition of the ashes.
** PGMG – polyhexamethylene guanidine chloride.

by impregnation and placed in containers of different sizes and configurations.

10.6 The state system for the accounting and control of RAW and radioactive materials (RAM)

The improvement and development of the state system for the accounting and control of radioactive material (RAM) helps to prevent the unlawful use of such materials and to facilitate radiation and environmental safety [27]. The national system of state accounting and control of RAW and RAM (NSSRMWAC) was created in accordance with Federal Law No. 170-FZ 'Nuclear Power Use' in November 1995. The purpose of the system is to register the existing quantities of RAM and RAW in their storage and disposal locations, to prevent loss, unauthorized use or theft of RAM and RAW, and to provide the national authorities and nuclear safety regulators with information regarding the availability, movements, exports and imports of RAW and RAM.

As per the Russian Government Decree No. 1298 of October 1997 on the Approval of Rules for the Organization of a National System of Accounting for Radioactive Materials and Wastes, the Russian Ministry of



(a)

(b)

(c)

(d)

(e)

10.9 Cementation by ash residue impregnation in 200 l barrel: (a) experimental-industrial installation, (b) initial radioactive ashes, (c, d) container and probe, (e) end product (cement compound with filling of the ash residue to 70% by mass).

Atomic Energy (Minatom, later Rosatom) was appointed responsible for all activities associated with the functioning of the accounting system for RAM and RAW. During 1998–2000, Minatom prepared the key legal acts and procedural documents to ensure the successful creation and operation of this system, including:

- the Statute of State Accounting and Control of RAM and RAW in the Russian Federation,
- procedural recommendations for primary inventory checks of RAW and RAM,
- forms for the provision of accounting data relating to RAW and RAM, along with instructions for the completion of these forms,
- forms for federal statistic supervision 2-TP (for radioactive materials), 2 TP (for radioactivity) along with recommendations for completion.

This documentation allowed a primary inventory of RAW and RAM to be introduced on 1 July 2000, with information starting to be collected from sites and analysed since 1 January 2001. Functionally, the organization of the national system of RAW and RAM accounting and control (Fig. 10.10) includes three levels of authority: federal (Rosatom), regional (regional executive authorities) and departmental (federal executive authorities). Under this system, Rosatom acts both as a federal and a departmental authority.

10.10 Organizational functional diagram of NSSRMWAC.

The federal level of the accounting system shall provide:

- federal-level accounting of RAW and RAM;
- collection and analysis of RAW and RAM accounting information at the regional and departmental levels;
- formation of databases for the state cadastre of RAW, RAW storage and disposal sites, and contaminated territories that are within the area of responsibility of the supervising organization;
- organization of information exchange between the authorities that control the accounting system at the federal, regional and departmental levels;
- development of scientific, procedural and technical developments that help create, operate and enhance the accounting system, providing the results to organizations involved with RAW and RAM control accounting at all levels;
- development of regulations and code documents (standard accounting forms for radioactive materials and waste, quantity and radionuclide composition measurement procedures, etc.), and, together with other federal executive stakeholders, of inter-compatible software for the databases;
- co-ordination of federal-level efforts;
- information for the federal authorities and nuclear regulators and other stakeholders regarding the availability, movement, imports and exports of radioactive materials and waste as required for these bodies to exercise their authority;
- management of the Information and Analytic Centre of the National System of Accounting and Control of Radioactive Materials and Radioactive Waste, providing information and analysis that helps the system to function at its federal level;
- co-operation with foreign nations on issues regulated by international agreements and programmes (projects) related to accounting of RAW and RAM.

The regional and departmental levels of the system perform similar functions scaled down to their areas of responsibility. Overall supervision of the system is entrusted to the Federal Service of the Ecological, Technical and Atomic Supervision of Russia (Rostechnadzor), which also licenses the corresponding forms of activity, and controls the observance of the established standards and rules for radioactive materials and waste management.

The system handles three flows of information. Information is supplied by every site, organization or subsidiary within 10 days of any operation involving the movement or change of status of RAM and RAW at its present location, transfer to other sites or legal persons, or receipt of RAM

and RAW. This ensures that radioactive substances are tracked throughout the entire management process, from the time of generation through all movements between sites and enterprises up to their classification as RAW. Similarly, movements and transitions of RAW are also tracked through to their placement in long-term storage or disposal locations. As the movement of RAM and RAW requires that reports be submitted by both the transferring and the receiving party, the security of the RAM and RAW during transfers between legal bodies is ensured.

Annual reporting carried out by sites and enterprises using the forms supplied by the federal statistics services not only helps to monitor whether operations involving RAM and RAW are being reported correctly, but also further analyses additional information about radioactive releases and effluents, contaminated land, and so on. The periodic taking of inventories (performed annually for RAM and once every five years for RAW) helps in the generation of accurate data regarding the availability and characteristics of these materials at their storage locations, as well as collecting additional information about the characteristics of the storage locations.

The creation and operation of this system has vastly improved the accounting of RAM and RAW at Russian sites. Another important factor in this improvement was also played by the introduction of the federal regulation Main Rules of Accounting for Radioactive Materials and Waste in Organizations [28] in 2006; increased supervision by Rostechnadzor with regard to the observance of these rules also played a key role. The results of the analysis of the information stored in the national system of accounting for radioactive materials and waste have been used for the production of a range of codes and guide documents, the most important of which is the Federal Program 'Assurance of Nuclear and Radiation Safety for 2008 and until 2015' and the Federal Law 'On Radioactive Waste Management'.

10.6.1 Norms and legislation governing RAW management in Russia

The various relevant aspects of activity are separately regulated at the legislative level by federal laws such as 'On the Use of Atomic Energy', 'On the Radiation Safety of the Population', and 'On the Sanitary-epidemiological Prosperity of the Population', among others. Russia's ratification of the united convention on the safe management of RAW and spent fuel shows that there is a general trend towards the creation and further development of the national normative lawful regulation of activity with regard to RAW management.

RAW treatment is currently regulated by the following standards and rules:

- 'Safety regulations regarding the rotation with radioactive wastes of atomic stations' (NP-002-04),
- 'Collection, processing, storage and conditioning of liquid radioactive wastes. Requirements of safety' (NP-019-2000),
- 'Collection, processing, storage and conditioning of solid radioactive wastes. Safety requirements' (NP-020-2000),
- 'Handling gaseous radioactive wastes. Safety requirements' (NP-021-2000),
- 'Health regulations for treatment with radioactive wastes' (SPORO-2002).

The regulations in these documents are applicable to all nuclear facilities, radiation sources and RAW processing units, whether planned, in preparation or operational. The set of documents listed above corresponds to the IAEA's recommendations concerning the regulation of RAW management.

The transportation of RAW and RAM is governed by health and safety regulations regarding:

- the transportation of radioactive materials (NP-053-04),
- RAW treatment (SPORO-2002),
- The radiation safety of staff and population during transportation of RAM (substances) (SP 2.6.1.128 1-03).

These documents present the established principles of RAW and RAM transportation, and the requirements put in place to ensure the safe transport of RAW and RAM.

RAW storage is regulated by:

- 'Rules on safety provision during the temporary storage of radioactive wastes, which are formed during the output, processing and use of minerals' (NP-052-04),
- 'Collection, processing, storage and conditioning of LRAW. Safety Requirements' (NP-019-2000),
- 'Collection, processing, storage and conditioning of SRAW. Safety Requirements' (NP-020-2000),
- Health regulations regarding RAW treatment (SPORO-2002).

Safety must be ensured during RAW storage in order to prevent staff, the general population and the environment from being exposed to radiation over the established limits both under normal operating conditions and in emergencies.

At the RAW disposal stage, safety measures aim to ensure reliable isolation of RAW, which in turn ensures radiation safety of the population and the environment for the whole period during which the RAW poses a potential hazard. The principal regulations regarding RAW disposal are:

- 'Radioactive waste disposal. Principles, criteria and basic safety requirements' (NP-055-04),
- 'Near-surface RAW disposal. Safety requirements' (NP-069-06),
- 'Recommendations regarding the establishment of the criteria of the acceptability of conditioned RAW for their storage and disposal' ([RB]-023-02),
- 'The safety evaluation of the near-surface repositories of radioactive wastes' (RB-011-2000);
- Health regulations regarding RAW treatment (SPORO-2002).

These documents establish the principles, criteria and basic safety requirements relating to near-surface RAW disposal, disposal into deep geological formations, and also for LRAW disposal. They establish a classification of near-surface disposals for RAW and make recommendations regarding safety evaluation methods for near-surface repositories. The regulations treat the methods used for RAW conditioning as a basic step in the preparation of RAW for storage and disposal.

10.6.2 New federal law 'On the Management of Radioactive Waste'

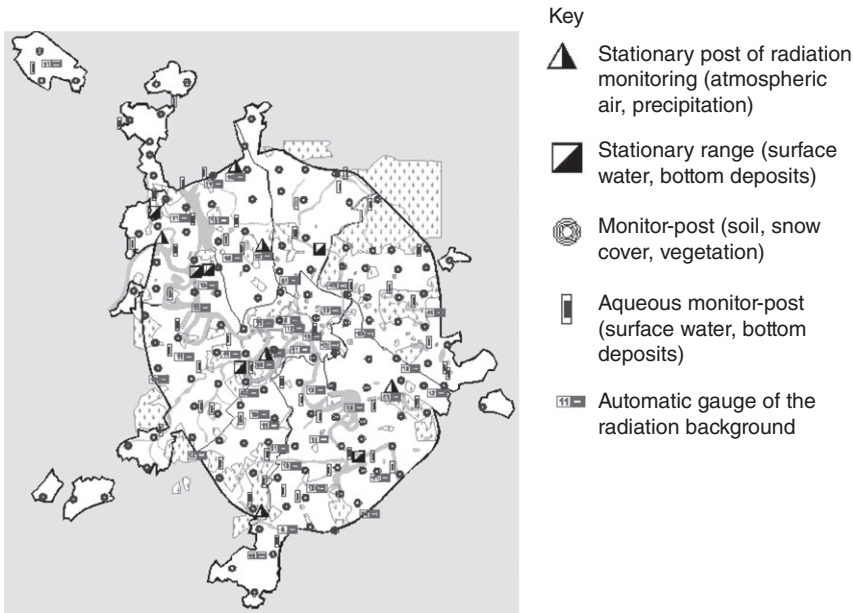
On 29 June 2011, the State Representative Assembly of the Russian Federation passed a law called 'On the Management of Radioactive Waste'. This federal law deals with crossover issues such as interim storage, the final isolation of RAW, and the financing of the measures required in RAW management. Crucially, it also lays out the responsibilities of the different authorities, including the Russian government, Rosatom, federal bodies, state and local government authorities, the national RAW management operator, and RAW suppliers. The law establishes the legal organizational basis for all forms of RAW management, as well as government control of RAW rates and costs and financial schemes. The requirements for RAW disposal, and the state's role in accounting, storage, control and registration are all determined by this law, along with the establishment of radiological controls and radiation monitoring. It further states that RAW containing nuclear materials is exclusively federal property. The system of state accounting and control of RAW and RAM is responsible for all RAW found on Russian territory, including registering RAW and its storage locations. Responsibility for newly generated RAW lies with the organization in which the RAW is produced: the same operations also take responsibility for the safe handling of RAW treatment up to the point of transfer to the national operator.

10.6.3 Radiation monitoring

Moscow has the world's first system of radiation-ecological monitoring (REM) [29], on the basis of observations taken of the environmental radiation characteristics of an area (Fig. 10.11).

The introduction of an analytical REM system [30] using information from radio-ecological data processing units has helped to solve the problems posed by integrated radio-ecological data processing. The system collects and analyses data from a variety of sources, including the subdivisions of the radiation emergency service, radiation-hygienic control, regional systems of RAW and RAM accounting, laboratory complexes, automated radiological control systems, and systems for monitoring the radon content in public buildings.

Studies on the detection of centres of radioactive contamination were all carried out in one facility in Moscow, which carried out accounting and monitoring of RAM and RAW and analysed the information that indicated the level of radiation present in the city, thereby ensuring the radiation safety of the population. A number of other projects were completed in the same facility, with the aim of providing radiation safety from natural radionuclides, by means of observation and control of natural sources of irradiation – (housing, industrial buildings, construction sites, and so on). Radiation-ecological monitoring of the environment was carried out



10.11 Layout of Moscow's REM locations.

in annual cycles by means of a system of stationary locations for monitoring radiation levels in the ground, air and water, while the individual radiation doses of Moscow citizens were also analysed.

Overall, a number of measures aimed at ensuring the population's radiation safety during the removal of RAW from the city were successfully developed. The work carried out in the REM framework 8 led to the acquisition of significant experience in developing radiation monitoring systems, which in turn has led to the optimization of the organization of environmental radiation control [31].

10.6.4 Advances in analytical control

Liquid scintillation (LS) spectrometry is the most widely used measurement method, and allows the simultaneous identification of beta-, alpha- and gamma-ray emitters, the decay schemes of which include the emission of low-energy electrons. A great deal of positive practical experience has been gained in the use of LS instruments for monitoring technological RAW and environmental samples of different composition and origin [32, 33]. The method is also sensitive enough to allow the rapid analysis of drinking water.

LS spectrometry is a rapid and simple alternative to labour-intensive alpha-spectrometric methods when specific isotopic analysis is required. With a minimum sample preparation stage, the radionuclide activity values can be obtained immediately after measurements are carried out (as with gamma-spectrometry) or, at least, it can be reliably determined whether the required levels for specific radionuclides have been exceeded. The spectrum obtained is processed with the help of RadSpectraDec software (Plate III (between pages 448 and 449)).

LS spectrometric analysis is used in two principal applications:

1. as part of RAW management technology for monitoring the radionuclide content by the screening method, without radiochemical preparation;
2. in radio-ecological studies, such as: monitoring of natural (Ra, Rn, U, Th, ^{210}Pb, ^{210}Po) and industrial (^{3}H, ^{90}Sr, ^{241}Pu) radionuclides in the environment at background levels, using radiochemical preparation of samples; and control of industrial radionuclides (^{3}H, ^{85}Kr, ^{89}Sr, ^{90}Sr, ^{99}Tc, ^{129}I, ^{234}U, ^{238}U, ^{241}Pu) in releases and discharges from facilities, using both the rapid method and the radiochemical preparation of samples.

10.6.5 Projects for the remediation of contaminated territories

1. In 2002, a project was initiated at the Kurchatov Institute with the aim of remediating contaminated objects and sites [34]. The principal tasks

of this project were the removal of the accumulated RAW, the decommissioning of old repositories and the remediation of the contaminated territory. The volumes involved were evaluated at 4,000 m^3 of SRAW, 20,000 m^3 of contaminated soils, and RAW activity of up to 700 Ci. In 2006, the decontamination of 10 old repositories was completed. More than 3,400 m^3 of SRAW with activity >500 Ci was removed: of this, over 3,000 m^3 was removed for long-term storage at MosNPO 'Radon', and 300 tons of metal low-level waste were sent for further melting at the 'Ekomet-S' facility.

2. A project to examine and decontaminate the buildings of Moscow's 'Zavod Polimetallov' plant was carried out between 1999 and 2003. There were 32 buildings and more than 9,000 m^2 of land. Decontamination of 17 buildings revealed an RAW volume of over 400 m^3, with radionuclides composed of ^{232}Th and ^{226}Ra.
3. The radiochemical laboratory of the Vernadsky Institute of Geochemistry and Analytical Chemistry (Russian Academy of Sciences) was in use from 1966 until the end of the 1980s. The contaminated objects at this site were seven hot cells with an operating area of 240 m^2, as well as 15 auxiliary premises, and a ventilation system. After a partial decontamination and dismantling of the equipment, it was decided to preserve the premises for 50 years.
4. The JSC 'Koltsugtsvetmet' (Vladimir Region) contained a workshop for the production of luminescent substances based on soluble ^{226}Ra bromides. This facility was built and put into operation in the mid-1950s. The two-storey building with a total area of 1,200 m^2 was decontaminated; the building and ventilation system were then dismantled and the surrounding territory remediated.

10.6.6 Establishment of the unified state system of RAW management: the FSUE 'RosRAO'

Prior to 2008, the united political system of the state management of RAW had to manage not only its basic role of developing state policy in the field of providing nuclear and radiation safety, but also a new and crucial function of solving the broader problems of the entire nuclear system (both historical and contemporary).

In 2008, a presidential decree ('On the Measures for the Creation of the 'Rosatom' State Corporation for Atomic Energy') determined that Radon, the enterprise with special responsibility for collection, storage and processing of RAW across Russia should be transferred to Rosatom. The reorganization of these enterprises on the basis of their relationship to the FSUE 'RosRAO' is now complete.

RosRAO was given the responsibility for managing the centralized collection and sorting of low and intermediate-level RAW, as well as their transport, conditioning and storage. The scope of the enterprise includes activities aimed at planning RAW volumes, developing technologies for RAW processing, designing and building units for final isolation, operating storage facilities, and remediating and monitoring, territories that were subjected to radioactive contamination. The law 'About the management of radioactive wastes management' gave RosRAO the functions of the national operator, and it now manages all the work and projects connected with solving the problems of accumulated RAW and other historical problems across the whole of the former USSR.

The Kirovo-Chepetsk Chemical Enterprise has been producing uranium fluorides since 1958. The area of contaminated facilities includes more than 100,000 m^3 of buildings and sludge repositories containing RAW with an activity of more than 3,000 Ci. The most basic form of decommissioning, preservation, has been carried out here: i.e., accumulated RAW remains on the territory of the Kirovo-Chepetsk department of FSUE 'RosRAO'. The preparation of the site for decommissioning requires the development of a decommissioning plan, the completion of scientific research, and the carrying out of crucial measures to guarantee safety. Full decommissioning will solve the problem of the existing RAW storage facilities, will remove contaminated buildings, and will create a new RAW storage site [35].

The remediation of sludge stores at the concentration unit of the former Novotroitsk mining plant constitutes another similar problem. The contaminated area there is more than 9 hectares in size, with over 200,000 m^3 of RAW (principally composed of ^{232}Th with 20 Ci of activity). The purpose of recent work relating to this site includes the preparation of technical and up-to-date documentation to allow remediation of the contaminated site and the transfer of both the technological equipment and RAW in an ecologically safe state. The average specific activity of the ^{232}Th in the RAW at this site is 72 Bq/kg, ^{226}Ra – 26 Bq/kg, ^{40}K – 1100 Bq/kg. In the evaluation of the physical volumes and total activity of RAW, only the anomalies and the sites with known radioactive contamination were examined. The specific activity of ^{232}Th is equal to 780 Bq/kg, which exceeds the levels judged to be minimally significant. Activities at this level are encountered in areas with irradiation fields of above 1.0 μSv/hour.

The results taken in the contaminated territory during the 2009 investigation [36] identified 18 anomalies with an average specific activity 2,800 Bq/kg. The total volume of the contaminated constructions due to be dismantled was about 1,500 m^3.

10.7 Conclusion

This chapter presents the author's perspective on the principal aspects of RAW management in Russia. The problems connected with special features of radioactive waste accumulation in the former USSR were examined, with the Eastern Ural Track used as an example of the ecological threat posed by mistakes made during RAW processing.

A number of issues connected with the formation of the institutional RAW management system in the former USSR are discussed, including the establishment of the Radon system, the long-term isolation strategy, the different sorts of historical repository, and the restoration process for those repositories. The procedure for converting historical repositories into new long-term RAW storage repositories is also outlined.

A survey of contemporary technologies used in RAW management was carried out, including combined liquid RAW treatment, vitrification and plasma treatment. A large part of the chapter is dedicated to RAW cementation technology as the basic industrial method of low- and intermediate-level LRAW conditioning. This technology requires low capital investment and operational expenditures and satisfies the quality requirements of the Russian Standard. The creation of the state system of RAW and RAM accounting and control is discussed in detail, along with broad coverage of questions concerning the legal aspects of RAW management in Russia. Moreover, the key elements of the new federal law 'On the Management of Radioactive Waste' are presented, with particular reference to aspects affecting the construction and development of contemporary RAW management system.

A brief overview of territorial radiation monitoring and achievements in analytical control was provided. It should be noted that a multilevel system of radiation monitoring was organized in Moscow immediately after the events at Chernobyl NPP. The enormous volume of work on environmental radioactivity led to the development of analytical techniques such as the use of the liquid scintillation spectrometry method. The final part of the chapter focused on questions connected with the problem of the remediation of territories contaminated with radionuclides.

This chapter has therefore demonstrated that Russia possesses the complete spectrum of activities and systems connected with the management of RAW, including treatment of spent fuel, nuclear fuel cycle, decommissioning liabilities and durable long-term storage of conditioned RAW.

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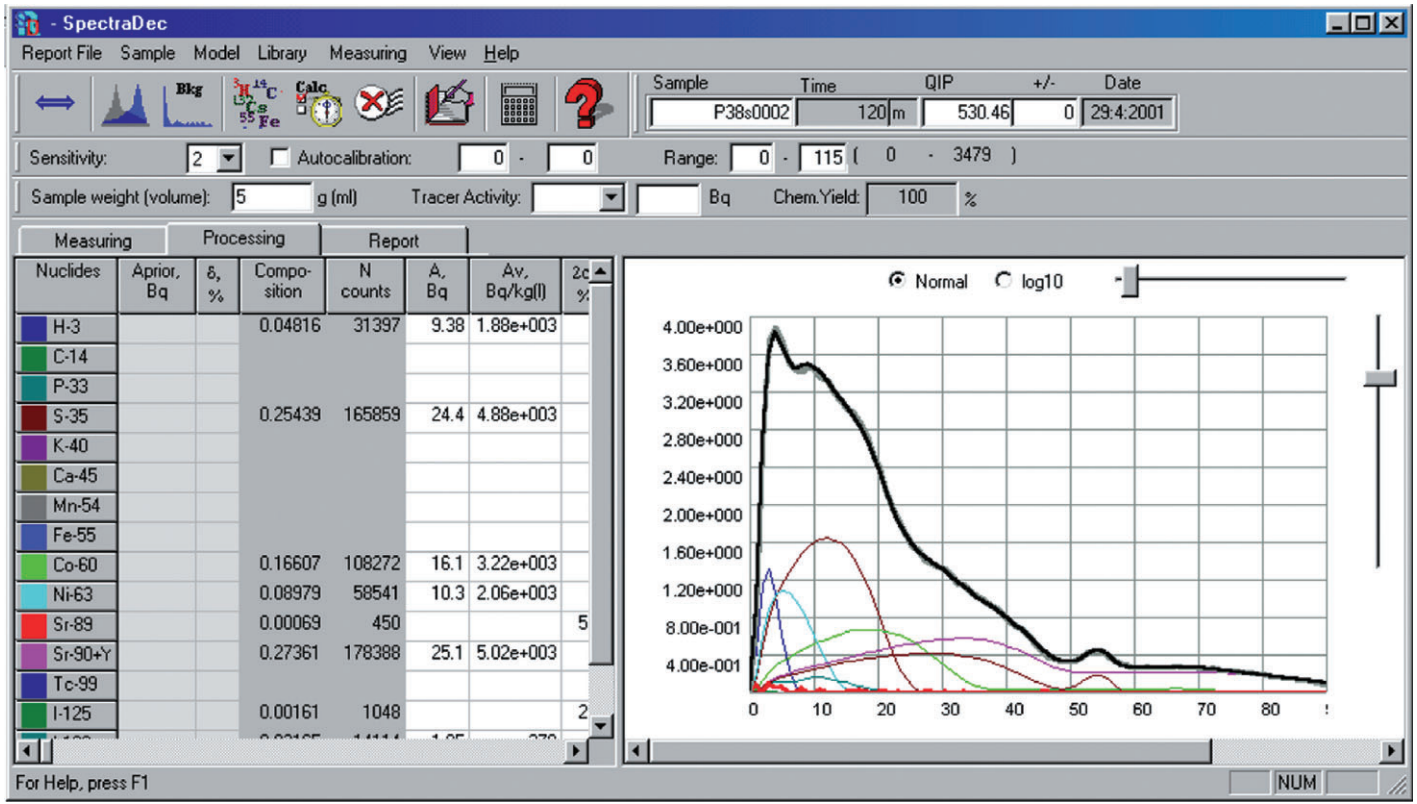

Plate III (Chapter 10) RadSpectraDec software interactive window.

11

Ukraine: experience of radioactive waste (RAW) management and contaminated site clean-up

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Abstract: A succinct description of the experience of radioactive waste management in Ukraine, which accumulated when using nuclear energy and developed as a result of the accident at Chernobyl nuclear power plant, is given in this chapter. In addition, the chapter summarizes the current state of the Chernobyl area contamination and the experience of Ukraine in decontamination and clean-up of the contaminated territories.

Key words: radioactive waste management in Ukraine, Chernobyl accident, decontamination and clean-up.

11.1 Introduction

This chapter describes the current state of radioactive waste (RAW) management experience in Ukraine, which resulted from its nuclear energy programme, and developed following the accident at the Chernobyl nuclear power plant (NPP). In addition, the current state of the Chernobyl area contamination and the experience of Ukraine in decontamination and clean-up of the contaminated territories is summarized.

Section 11.2 describes the main sources of radioactive waste in Ukraine and their current classification, and summarizes the information about the properties and amounts of accumulated waste. Section 11.3 describes radioactive waste management practices in Ukraine, including a brief summary of the legislation, responsibilities of various organizations involved, technical policy, and funding of waste management in Ukraine. Section 11.4 gives a concise description of the environmental consequences of the Chernobyl disaster, Ukraine's experience in decontamination and clean-up of the contaminated territories, and the current state of radioactive waste management in the Chernobyl Exclusion Zone. Section 11.5 sets out main modern organizational and technical problems in the field of radioactive waste (RAW) management. A description of Ukraine's recent initiatives aimed at solving the existing problems is given in Section 11.6. Sections 11.7 and

11.8 contain conclusions and a description of additional sources of information.

It should be emphasized that in preparing this chapter, the information regarding the quantity of radioactive waste and waste management technologies was taken from official sources (National Reports of Ukraine, normative documents, etc.). As a result, the information may differ slightly from the data contained in sources of information published after 2009. This is particularly true regarding RAW from NPP.

11.2 Sources, types and classification of wastes

11.2.1 Sources of RAW in Ukraine

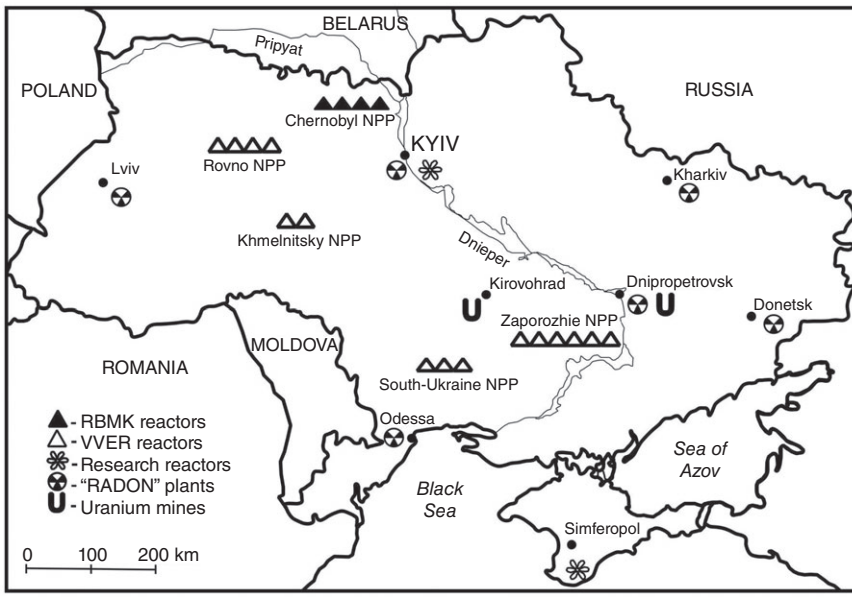
Ukraine has been using nuclear technologies intensively now for several decades. Current Ukrainian nuclear energy policy is directed towards an increase in nuclear power development. Thus, radioactive waste management is, and will remain, an important safety issue in Ukraine.

Radioactive waste is generated at operating NPP, at other facilities of the nuclear fuel cycle, as well as outside the nuclear fuel cycle (e.g., at research reactors, from non-nuclear industries, in medical and scientific institutions, where radiation sources are also used). Some legacy RAW is stored at facilities of the Ministry of Defence and Border Service. Significant amounts of RAW resulted from the 1986 reactor accident at unit 4 of Chernobyl NPP (ChNPP). In the near future, further radioactive waste generation is expected as a result of the decommissioning of the ChNPP, other nuclear reactors and facilities, as well as from the clean-up of radioactively contaminated sites.

Nuclear energy in Ukraine started to develop in the 1970s with the construction of the first 1000 MW(e) RBMK nuclear reactor at Chernobyl. Currently, 15 reactors are being operated at four NPP (13 VVER-1000 reactors and 2 VVER-440 reactors). Four RBMK reactors were constructed at Chernobyl, one of which was destroyed in the 1986 disaster, while the other three units are now shut down. Ukrainian NPPs (Fig. 11.1) generate almost half of the national electricity production (up to 48%). The total installed capacity of the Ukrainian operating units is 13.8 GW. Ukraine ranks 7th in the world and the 5th in Europe in terms of installed nuclear capacity. In 2009, Ukrainian NPP produced 83.2×10^9 kW·h of electricity.

11.2.2 Current classification of RAW

The existing waste classification system in Ukraine has been derived on the basis of regulatory documents (RSNU, 1997; ADD, 2000; MSRU, 2005). RAW in Ukraine is subdivided into types, groups, categories, and kinds



11.1 Location of Ukrainian nuclear facilities.

depending on the classification purpose. The following are key elements of the classification system:

- subdivision into solid and liquid waste,
- classification based on the generic waste acceptance criteria for radioactive waste disposal in different repository types; this classification includes short-lived waste, for disposal in near-surface or surface repositories, and long-lived waste, for disposal in a geological repository (Table 11.1),
- classification of solid radioactive waste by the 'exemption level' criterion (sec. 15.1.6 from MSRU, 2005),
- classification of solid radioactive waste (SRAW) and liquid radioactive waste (LRAW) by specific activity (Table 11.2),
- classification of RAW with unknown radionuclide composition according to the absorbed radiation dose rate in air at 0.1 m distance from the object (container) surface (sec. 15.1.8 from MSRU, 2005).

According to Ukrainian legislation, spent nuclear fuel (SNF) is not declared as radioactive waste. According to *Radiation Safety Norms of Ukraine* (RSNU, 1997) waste from uranium mining processing, as well as waste resulting from mining other mineral products, is related to technologically enhanced naturally occurring radioactive materials (TE-NORM).

Table 11.1 Waste classification based on the criteria of acceptance for disposal in different types of facilities

Waste type	Potential radiation dose rate in 300 years after the waste disposal	Type of possible clearance from regulatory control during the period less than 300 years after the waste disposal	Allowed waste disposal facility type
Short-lived	Below 1 mSv·yr^{-1}	Complete, limited	Surface or near-surface
Long-lived	Above 50 mSv·yr^{-1}	Not considered	In stable deep geological formations

Table 11.2 Classification of solid and liquid waste by specific activity

Category	Waste group				Liquid RAW specific activity as a multiplier of PC_B^{ingest} units[a]
	1	2	3	4	
	Solid RAW specific activity range (kBq·kg^{-1})				
Low level waste (LLW)	10^{-1}–10^{1}	10^{0}–10^{2}	10^{1}–10^{3}	10^{2}–10^{4}	1–10^{2}
Intermediate level waste (ILW)	10^{1}–10^{5}	10^{2}–10^{6}	10^{3}–10^{67}	10^{4}–10^{8}	10^{2}–10^{6}
High level waste (HLW)[b]	≥10^{5}	≥10^{6}	≥10^{7}	≥10^{8}	≥10^{6}

[a] PC_B^{ingest}: permissible level that limits specific volumetric activity of radionuclides in drinking water according to RSNU (1997).
[b] The HLW category is subdivided into two subcategories: 'low temperature' HLW with specific heat generation at the temporary storage or final disposal places not exceeding 2 kW·m^{-3}, and 'heat generating' HLW with specific heat generation of more than 2 kW·m^{-3}.

These kinds of radioactive materials are also not declared as radioactive waste.

11.2.3 Inventory of accumulated waste

RAW of nuclear energy sector

NPPs are currently the main radioactive waste producers in the Ukraine, producing liquid and solid RAW. The main sources of primary liquid

radioactive waste (LRAW) from NPPs are pipelines leakages, water from spent nuclear fuel storage pools, solutions remaining after sorbent regeneration and spent decontamination solutions. The product of primary LRAW reprocessing (except for spent filtering materials and sludge) is concentrated salt solutions, which are exposed to deep evaporation, resulting in a fusion cake.

The volume of LRAW at Ukrainian NPPs per 10^9 kW h of the generated electricity is evaluated to be (Shestopalov *et al.*, 2008):

- evaporation bottoms: about 13 m^3
- filtering materials and sludge: about 8 m^3
- fusion cake: about 2.4 m^3
- oil and mixed solutes: 0.45 m^3

The isotope composition of LRAW is mostly: ^{90}Sr, ^{90}Y, ^{134}Cs, ^{137}Cs, which come from untight heat-emitting elements, and ^{58}Co, ^{60}Co, ^{54}Mn, ^{59}Fe, ^{51}Cr, ^{124}Sb, which are formed by neutron activation of pipes and contour corrosion products.

Specific activities of LRAW are within the following range:

- evaporation bottoms and fusion cake: from 10^{10} to 10^{11} Bq m^{-3};
- filtering materials and sludge: from 10^9 to 10^{10} Bq m^{-3}.

Table 11.3 presents the data related to LRAW accumulated at Ukrainian NPPs as of mid-2008 (NatRep, 2008).

The sources of solid radioactive waste (SRAW) from NPPs are worn-out equipment, apparatus and instruments; dismantled equipment and pipelines, construction materials and debris; used individual protection means; elastron, electric- and heat insulation materials; ventilation system spent filters, and sludge from treatment facilities. In addition, NPPs store spent ionizing radiation sources.

The isotope composition of SRAW waste is mostly: ^{137}Cs, ^{134}Cs, ^{60}Co, ^{90}Sr, ^{95}Nb, ^{54}Mn, ^{51}Cr, ^{59}Fe.

Table 11.3 Liquid radioactive waste at Ukrainian NPPs as of mid-2008

NPP	Evaporation bottoms (m^3)	Filtering materials (m^3)	Fusion cake (m^3)
Zaporozhie	2,100	350	5,400
South Ukraine	2,850	230	–
Rovno	5,160	525	770
Khmelnitsky	450	155	780
Total	10,670	1,260	6,950

The volume of SRAW at Ukrainian NPPs per 10^9 kW h of the generated electricity is evaluated to be (Shestopalov *et al.*, 2008):

- low level waste (LLW): about 30 m^3
- intermediate level waste (ILW): about 1.0 m^3
- high level waste (HLW): about 0.1 m^3

Table 11.4 presents the data related to SRAW accumulated at Ukrainian NPPs as of mid-2008 (NatRep, 2008).

Non-nuclear sector RAW

The State Interregional Specialized Plants (SISP) of the Ukrainian State Association 'Radon' (UkrSA 'Radon') deal with collection, transportation, storage and disposal of RAW from Ukrainian enterprises, medical and research institutions, including ionizing radiation sources. SISP are located near Dnipropetrovsk, Kyiv, Lviv, Odessa and Kharkiv (Fig.11.1).

Table 11.5 presents the data related to radioactive waste accumulated at SISP of the UkrSA 'Radon' as of mid-2008 (NatRep, 2008).

Two research reactors in Ukraine (Fig. 11.1) store liquid and solid radioactive wastes on site:

- reactor WWR-M of the Nuclear Research Institute of the National Academy of Sciences of Ukraine (Kyiv);
- reactor IR-100 of the National Institute for Nuclear Energy and Industry (Sevastopol).

RAW in the Chernobyl Exclusion Zone

As a result of the accident at unit 4 of ChNPP, a large quantity of radioactive material was released and distributed over a huge territory. Most of these materials remain inside the unit above which the 'Shelter' object has been built, and within a local area (the so-called 'Shelter' object site) that surrounds the ruined unit and ChNPP site. According to a first estimate, the

Table 11.4 Solid radioactive waste at Ukrainian NPPs as of mid-2008

NPP	LLW (m^3)	ILW (m^3)	HLW (m^3)
Zaporozhie	7,620	620	87
South-Ukraine	15,980	565	13
Rovno	5,780	1,020	63
Khmelnitsky	3,890	110	9
Total	33,270	2,315	172

Table 11.5 Radioactive waste at UkrSA 'Radon' plants as of mid-2008

SISP location	Amount (m^3)	Weight (t)	Activity (Bq)	Main radionuclides
Low and intermediate level solid waste				
Kyiv	1,994	2,715	5.8×10^{15}	^{137}Cs, ^{239}Pu, ^{60}Co, ^{3}H, $^{90}Sr + {}^{90}Y$
Dnipropetrovsk	433	1,101	6.0×10^{15}	^{137}Cs, ^{192}Ir, ^{239}Pu, ^{3}H, ^{226}Ra
Odessa	403	314	5.1×10^{14}	^{60}Co, ^{137}Cs, ^{239}Pu, ^{3}H, $^{90}Sr + {}^{90}Y$
Lviv	640	1,513	4.1×10^{12}	^{137}Cs, ^{239}Pu, ^{60}Co, ^{3}H
Kharkiv	1,633	2,438	9.6×10^{12}	^{137}Cs, ^{239}Pu, ^{60}Co, $^{90}Sr + {}^{90}Y$
Low and intermediate level liquid waste				
Kyiv	413	413	1.8×10^{12}	^{137}Cs, ^{60}Co, ^{3}H, ^{192}Ir
Dnipropetrovsk	60	60	1.7×10^{10}	^{137}Cs, ^{60}Co, ^{3}H
Odessa	138	138	1.1×10^{11}	^{14}C, ^{3}H
Kharkiv	10	10	2.3×10^{8}	^{137}Cs, ^{60}Co, ^{3}H
Spent sealed radiation sources				
Kyiv	–	–	7.2×10^{14}	^{137}Cs, ^{60}Co, ^{3}H, ^{192}Ir
Dnipropetrovsk	–	–	4.5×10^{14}	^{137}Cs, ^{226}Ra, ^{60}Co, ^{192}Ir
Odessa	–	–	1.5×10^{14}	^{137}Cs, ^{226}Ra, ^{60}Co, ^{192}Ir
Lviv	–	–	3.6×10^{14}	^{137}Cs, ^{60}Co, ^{192}Ir
Kharkiv	–	–	3.5×10^{14}	^{137}Cs, ^{60}Co, ^{192}Ir
Spent sealed radiation sources of high power (RITEG)				
Odessa	–	–	2.99×10^{16}	$^{90}Sr + {}^{90}Y$

nuclide composition of this radioactive waste corresponds to the nuclide composition of irradiated nuclear fuel of RBMK-1000 reactors with an average burn-up of 11,000 MW day t^{-1}. At this rate, the ratio of activities of gamma- and beta-emitting nuclides to alpha-emitting nuclides is approximately 100 to 1.

Two main radioactive waste sources can be defined in the Chernobyl Exclusion Zone (ChEZ):

- radioactive waste generated during the operation of the four ChNPP units (Table 11.6);
- radioactive waste resulting from the accident at unit 4. Accidental wastes are located in the 'Shelter' object (Table 11.7) and in radioactive waste disposal points (RWDP) and radioactive waste temporary storage points (RWTSP) (Table 11.8).

Table 11.6 Radioactive waste of ChNPP

Category	Amount (m^3)	Activity (Bq)	Main radionuclides
Solid radioactive waste			
LLW	1,069	1.1×10^{11}	Cs, Sr, Co, Pu, Am
ILW	926	4.1×10^{12}	Cs, Sr, Co, Pu, Am
HLW	507	1.3×10^{14}	^{137}Cs, ^{134}Cs, ^{60}Co, ^{94}Nb, ^{154}Eu, ^{152}Eu, ^{241}Am, ^{243}Am
Liquid radioactive waste			
LLW and ILW	20,260	3.8×10^{14}	^{137}Cs, ^{134}Cs, ^{60}Co, ^{90}Sr, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Am

Source: NatRep (2008).

Table 11.7 Radioactive wastes of the 'Shelter' object, its site, and the ChNPP site

Radioactive waste characteristics	Category	Amount	
		m^3	t
'Shelter' object			
Liquid waste – water in the 'Shelter' rooms	LLW and ILW	3,000	
Fuel-containing materials – pieces of the core and lava-like fuel containing materials	HLW		3,000
Containers with pieces of the core and others	HLW	1,700	
Construction materials	HLW		121,000
Construction materials	LLW and ILW		330,000
'Shelter' object industrial site			
Concrete, concrete plates and units	HLW	1,500	
Concrete, crushed stone, sand, gravel	LLW and ILW	280,000	
ChNPP industrial site			
Concrete, metal, soil	LLW and ILW	500,000	

Source: NatRep (2008).

The main characteristics of the 'Shelter' object (SO) RAW are given in Table 11.7. The total waste activity of the SO as of the beginning of 2005 is approximately 4.1×10^{17} Bq, and the waste volume (according to different estimates) is between 530,000 and 1,730,000 m^3.

The volume of waste concentrated in RWDP and the main RWTSP of the ChEZ is approximately 2 million m^3, and the total activity is estimated at 7.7×10^{15} Bq. It should also be noted that the same amount of radionuclides is again contained in natural objects (vegetation, soils, bottom

Table 11.8 Radioactive waste in RWDP and RWTSP of the exclusion zone

Title	Volume (m^3)	Weight (t)	Activity (Bq)
RWDP (mainly LLW and ILW)			
'Buriakovka'	590,000	1,120,000	2.7×10^{15}
'The III line of ChNPP'	26,200	41,900	3.6×10^{14}
'Podlesny'(LLW and ILW)	7,040	14,080	2.5×10^{12}
'Podlesny' (HLW)	3,960	7,920	2.6×10^{15}
RWTSP (LLW and ILW)			
'Staraya stroybaza'	171,000	316,000	1.1×10^{15}
'Novaya stroybaza'	150,000	70,000	1.9×10^{14}
'Ryzhy les'	500,000	250,000	3.7×10^{14}
'Neftebaza'	102,000	181,000	3.7×10^{13}
'Stantsiya Yanov'	30,000	15,000	3.7×10^{13}
'Kopachi'	110,000	90,000	3.3×10^{13}
'Pripyat'	16,000	11,000	2.6×10^{13}
'Peschanoye plato'	57,300	91,700	6.4×10^{12}
'Chistogalovka'	160,000	150,000	3.7×10^{12}

Source: NatRep (2008).

deposits, underground water) of the radioactively contaminated landscape. Table 11.8 gives the characteristics of waste in RWDP and the RWTSP.

Outside the ChEZ, in the Kyiv, Zhytomir, and Chernigov regions, there are several waste storage facilities built as a result of decommissioning and remediation activities in these areas. Their total volume of waste is 171,000 m^3 and the total activity of these materials can be between 1 and 2×10^9 Bq.

Total waste inventory

Table 11.9 summarizes the data on total amount of radioactive waste accumulated in Ukraine by the middle of 2008. In the future, new streams of radioactive waste will originate in Ukraine. It is assumed that in accordance with international agreements, vitrified high-level RAW will arrive to Ukraine from the Russian Federation starting in 2013. These wastes are the product of reprocessing and subsequent vitrification of spent nuclear fuel from Ukrainian VVER-440 reactors.

According to Shestopalov *et al.* (2008), in the course of decommissioning and dismantling of existing Ukrainian VVER reactors, the following amounts of reprocessed solid waste will appear (in thousand m^3): LLW: 34.4; ILW: 4.6; HLW: 2.4.

In the future, the rate of RAW accumulation in Ukraine will increase. This is due to Ukraine's plans to increase the annual electric power production at NPP from 88×10^9 kWh in 2005 to 220×10^9 kWh in 2030 (Energy Strategy, 2006).

Table 11.9 Radioactive waste accumulated in Ukraine as of mid-2008

Waste source	Amount (m^3)	
	Solid waste	Liquid waste
Operating NPPs	35,670	18,880
Chernobyl NPP	2,500	20,260
'Shelter' object	530,000–1,730,000	3,000
RWDP and RWTSP within the Chernobyl Exclusion Zone	1,913,000	–
RWTSP outside the Chernobyl Exclusion Zone	171,000	–
UkrSA 'Radon' plants	5,100	620
Research reactors	30	370

11.3 Radioactive waste (RAW) management practice

11.3.1 Legislation

After separation from the Soviet Union, Ukraine has implemented a well-developed legislation and regulatory framework for its nuclear industry. Radioactive waste management is carried out in accordance with the laws and other legal acts of Ukraine. These documents can be divided into three levels:

- international agreements, laws and resolutions of the government of Ukraine;
- the system of special rules and regulations to ensure safety during RAW management, which are based on the laws and establish the procedure for certain types of activities;
- operational documentation, instructions, procedures and regulations.

The basic laws of Ukraine, which regulate the activity for RAW management are:

- The law of Ukraine 'On the Use of Nuclear Energy and Radiation Safety' (1995).
- The law of Ukraine 'On Radioactive Waste Management' (1995).

11.3.2 National policy in the field of RAW management and responsibility of main 'players'

National policy in the field of RAW management

Major principles of state policy in the field of RAW management are stated in the laws of Ukraine 'On the Use of Nuclear Energy and Radiation Safety' and 'On Radioactive Waste Management'. They are as follows:

- priority to protection of personnel, population and environment against the impact of RAW;
- isolation of RAW from the environment;
- regulatory control of RAW management;
- separation of the regulatory functions and RAW management functions;
- responsibility of RAW producers for its safety;
- decisions on siting of new RAW management facilities taking into account society's opinions;
- prohibition of transfer to Ukraine of RAW for storage or disposal.

The principles of State policy in the field of spent fuel (SF) management are stated in the law of Ukraine 'On the Use of Nuclear Energy and Radiation Safety'. Energy Strategy (2006) defines the steps in the implementation of the so-called 'deferred' decision for SF of Ukrainian NPPs – long-term (up to 50 years and more) storage of SF with subsequent approval of final decision on either processing or disposal.

Main players and their responsibilities

In Ukraine, the following functions are differentiated at the legislative level:

- public administration in the field of nuclear energy use and governmental regulation of nuclear and radiation safety;
- public administration in the field of nuclear energy use and RAW disposal.

In Ukraine the competences and responsibilities for RAW management are distributed at the national level as follows.

- *Ukrainian Parliament (Verkhovna Rada)*: adoption of laws in the area of nuclear energy utilization and waste management including, when necessary, construction, design and siting of nuclear facilities and RAW management facilities of national importance (including near-surface disposal and disposal in geological formations).
- *Cabinet of Ministers*: decision making on construction, design and siting of nuclear installations and RAW management facilities with the exception of those under the competence of Parliament, management of nuclear facilities and RAW management facilities being the national property.
- *Ministry of Public Health*: establishment of radiation safety regulations, regulatory and supervisory activities over following norms and rules of radiation safety, issuance of sanitary passports for RAW management facilities.

- *State Nuclear Regulatory Inspectorate*: establishing normative criteria for following radiation safety norms, issuing licenses for implementation of activity on RAW management, supervision over compliance with regulatory requirements and conditions of license including coercive actions.
- *Ministry of Energy and Coal Industry*: national administration in the area of RAW management at Ukrainian NPPs, including co-ordination of activities on construction and operation of radioactive waste management facilities at NPPs.
- *Ministry of Emergency*: national administration in the sphere of RAW management, including co-ordination of activity on construction and operation of RAW disposal facilities.

Any practical activities are carried out by the State Specialized Companies (SSC) of the Ministry of Energy and Ministry of Emergency within their competence. Such companies shall have the licence for conducting activities on siting, designing, constructing, operating, decommissioning nuclear facilities or siting, designing, constructing, operating, and closing of a RAW storage facilities. They shall ensure nuclear and radiation safety and bear responsibility for nuclear damage.

Financing of RAW management

According to the Ukraine government strategy (Strategy, 2009), until recently the cost of RAW management at NPP was included in the price of electric energy. The following were funded from the State Budget of Ukraine:

- management of waste from research nuclear reactors;
- decommissioning of units 1–3 of Chernobyl NPP;
- RAW management at the SSE 'Complex' and the enterprises of UkrSA 'Radon';
- construction of the 'Vector' complex.

Work for decommissioning of Chernobyl NPP units 1–3 was also funded by international technical assistance programmes, and RAW management at the enterprises of UkrSA 'Radon' – partly at the expense of companies supplying RAW.

Budgetary funding was sufficient only to maintain the level of safety achieved for RAW management facilities. Implementation of modernization, reconstruction and construction of new storage facilities and other objects of RAW management infrastructure, as well as implementation of measures to improve safety was funded according to the residual principle. Reservation of funds for the forthcoming costs of RAW disposal was not carried out.

11.3.3 Current state of RAW management in Ukraine

RAW management at Ukrainian NPP

Storage facilities of standard design are built and operated at NPP sites for low-, intermediate- and high-level solid and liquid RAW as well as compaction, deep evaporation and decontamination facilities.

Liquid waste is treated at active water treatment systems, and then treated water returns to the technological process, and final treatment products (secondary waste such as evaporation bottoms, slurry, spent sorbents and salt fusion cake) are transferred to liquid waste storage facilities for cooling and temporary storage. For this purpose, NPPs use storage facilities in the form of iron tanks located in separate buildings.

To collect and temporarily store solid waste resulting from operation of NPP units, storage facilities located in separate buildings are used. Storage facilities are a series of concrete cells, which are lined with 4 mm stainless steel and equipped with biological protection. The thickness of the protection ranges from 600 to 1500 mm, depending on the specific activity of the waste. The depth of the cells ranges from 5 to 18 m. The waste is stored in bulk form in this type of storage facilities.

The following types of waste processing facilities are operated at Ukrainian NPPs:

- deep evaporator for processing of evaporation bottoms;
- solid waste incinerator for processing of solid low level radioactive waste;
- solid waste compactor unit for processing of solid low level radioactive waste;
- solid waste sorting unit for processing of solid low level radioactive waste;
- centrifugation system for processing of floor drains;
- radioactive oil incinerator.

However, each Ukrainian NPP is equipped with these facilities to different extents. Table 11.10 provides information on waste processing facilities, which operate at NPPs in Ukraine (SNRCU, 2009).

It is planned that a waste processing complex will be commissioned at Zaporozhie NPP. The incinerator, the compactor unit and the defragmentation unit are being designed. The Zaporozhie NPP solid waste storage facilities are 70% full. To extend the volume of the storage facilities by 30%, a new storage facility intended for storage of containers with salt fusion cake to be removed from storage facilities is being constructed.

At Rovno NPP, the following design activities of the solid waste processing plant are underway, in particular: retrieval facility, fragmentation and sorting facility, super-compactor and radiation monitoring system in waste management.

Table 11.10 Waste processing facilities at Ukrainian NPPs

Waste processing facility	Zaporozhie NPP	Rovno NPP	South Ukraine NPP	Khmelnitsky NPP
Evaporator	Yes (Two units)	Yes (Two units)	No	Yes
Incinerator	Yes	No	No	No
Compactor	Yes	No	Yes	No
Sorting unit	Yes	No	No	No
Centrifuge system	No	Yes	No	Yes
Oil incinerator	No	No	No	Yes

The following conclusions were drawn in SNRCU (2009), as a result of the safety assessment of waste management at NPPs:

- operating NPPs have technical capacities to temporarily store solid and liquid waste (also for extension of power unit operation lifetime) and to monitor its storage;
- the interaction with specialized enterprises dealing with disposal of waste should be expedited with regard to the waste acceptance criteria for storage or disposal.

RAW management in the non-nuclear sector

At the present time, the State Interregional Specialized Plants (SISP) of the UkrSA 'Radon' are intended mainly for non-fuel cycle waste storage. They carry out the following types of operation:

- storage of liquid and solid waste and spent ionizing radiation sources of industrial, medical, and scientific and research institutions;
- waste transportation, as well as spent ionizing radiation sources from their collection and temporary storage locations;
- elimination of possible radiological consequences of accidents which may be connected with lost radiation sources or legacy waste location.

Each SISP of the UkrSA 'Radon' operates vault-type storage facilities for solid radioactive waste, well-type storage facilities for ionizing radiation sources, and tank-type storage facilities for liquid waste.

The vault-type storage facilities are used for storage of SRAW in bulk form. They are a series of reinforced concrete cells. A separate cell has dimensions of $14.5 \times 9.5 \times 3.0$ m and capacity of 400 m^3. Several such storage facilities can be located on SISP sites. Well-type storage facilities for ionizing radiation sources are built as an underground tank which is connected

to the surface by a winding pipe 104 mm in diameter. The tank and the connective pipe are made of stainless steel and are concreted at a 6 m depth. The receiving device is provided with a plug and metal cover. Storage facilities for liquid radioactive waste are underground cylindrical tanks 9 m in diameter and 3.15 m in height. They are made of stainless steel and coated with reinforced concrete.

11.4 Chernobyl accident

11.4.1 Environment contamination from the accident and its current state

The extent of surface contamination

The 1986 accident at the Chernobyl nuclear power plant, resulted in a substantial release of radionuclides to the atmosphere and caused extensive contamination of the environment. According to the International Atomic Energy Agency (IAEA, 2006), a small part of the nuclear fuel (up to 3.5%) and a substantial fraction of volatile radionuclides were released from the damaged unit 4. The total activity amounted to approximately 12.5×10^{18} Bq, and included 6.5×10^{18} Bq of noble gases (IAEA, 2001).

A considerable territory of the former Soviet Union, particularly in Belarus, Russia, and Ukraine, as well of Western Europe, primarily the Scandinavian countries and the Alpine region, was severely contaminated. High levels of radioactive contamination in areas outside the Chernobyl Exclusion Zone arose for the following reasons: release of contaminated masses into the atmosphere to a height of 2,000 m and their intense movement at these altitudes; rainfall; and the presence of complex landscapes that dictated changes in directions and altitudes of the contaminated air masses movement.

The overall area of Western Europe countries where levels of ^{137}Cs contamination exceeded 20 kBq m^{-2} (almost 10 times higher that global background levels) due to the Chernobyl disaster amounted to approximately 280,000 km^2. Almost 75% of Ukraine's territory suffered from radioactive contamination by ^{137}Cs, which exceeded the pre-accident levels by more than double. The radionuclide decay, which has occurred in the 25 years since the Chernobyl accident, substantially corrected a pattern of radionuclide distribution over Ukraine's territory. Over this period, the area of localities where ^{137}Cs contamination levels exceeded 10 kBq m^{-2} has reduced to almost half what it was immediately after the accident. The area of sites where ^{90}Sr contamination exceeded 4 kBq m^{-2} is now less than one third, i.e. practically 90% of Ukraine's territory is characterized by the pre-accident levels of ^{90}Sr contamination.

However, the level and extent of Ukraine's territory contamination by Pu isotopes have not changed. ^{241}Am activity is gradually increasing due to ^{241}Pu decay; and the area of its distribution where levels exceed 0.2 kBq m^{-2} shall be 30% wider than the area of plutonium isotope fallout having the same density. The area of Ukraine contaminated by ^{90}Sr, ^{241}Am, and Pu isotopes is substantially smaller than that contaminated by ^{137}Cs.

Severely contaminated (over 1.5 MBq m^{-2} of ^{137}Cs) localities (almost 300 km^2) within the boundaries of ChEZ will remain uninhabitable for hundreds of years. These water-producing areas shall remain a long-term source of surface water and groundwater contamination due to surface washout and vertical migration.

Radioactive contamination of the air

Total β-activity of atmospheric aerosols increased rapidly in April 1986 as the result of radionuclide release from the damaged reactor. However, starting from 1989, the β-activity of aerosols has been primarily from naturally occurring radioactive elements.

In the contaminated areas, where farming activities (ploughing, harrowing, etc.) are actually not performed, the total β-activity is 2–3 times less than that observed in areas not classified as radioactive contaminated zones. In case of intense works involving destruction of a soil surface layer, where the ^{137}Cs contamination level is 370–555 kBq m^{-2}, the radionuclide volumetric activity in aerosols at a height of 3–5 m may exceed the limits established by RSNU (1997).

The present average annual concentrations of ^{137}Cs and ^{90}Sr in aerosols are similar to the pre-accident levels, i.e. 0.08×10^{-5} Bq m^{-3}.

Radioactive contamination of surface water

Following the Chernobyl accident (to 2000–2004), the ^{137}Cs content in most Ukrainian rivers (lower reservoirs on the Dnipro River, the Desna River and the Danube River) reduced to the pre-accident levels of 0.5–1.5 Bq m^{-3}, demonstrating the effects of natural attenuation processes such as radionuclide vertical migration into soil and its irreversible fixation by soil particles. Relatively high levels of radionuclide activity are still observed in the Pripyat River and in other waterways within the ChEZ (50–300 Bq m^{-3} for ^{90}Sr, 20–80 Bq m^{-3} for ^{137}Cs). At the same time, ^{90}Sr content in the Pripyat River is about 1.0–1.5 orders of magnitude lower than allowable levels of radionuclides in drinking water (2000 Bq m^{-3}) established in Ukraine.

During 2008–2009, the radionuclide concentrations in the water of the Kyiv Reservoir for ^{90}Sr ranged from 40 to 100 Bq m^{-3} and for ^{137}Cs from 10 to 20 Bq m^{-3}, while in the Kakhovka Reservoir (lowest in the cascade of

Dnieper reservoirs), the ^{137}Cs activities varied from 0.5 to 1.0 Bq m^{-3}. Radionuclide washout in the rivers is constantly decreasing. On a background of reduction of radionuclides by surface runoff, the annual infiltration outlet of contaminated water from the Chernobyl cooling pond remains more or less stable, making its relative contribution to the Pripyat River contamination higher compared to surface runoff.

Bottom sediment of the Kyiv Reservoir

The total inventory of ^{137}Cs and ^{90}Sr in the bottom sediment of the Kyiv Reservoir has gradually reduced since 1990. This is due to radioactive decay which dominates natural attenuation processes and surpasses the annual radionuclide intake associated with the river's runoff inlet and is also due to partial removal of radionuclides from the water to the bottom sediment due to adsorption sedimentation processes.

In the first decade following the accident, approximately 70% of ^{137}Cs and 90% of ^{90}Sr total intake to the Dnipro reservoir system was accumulated by the bottom sediment of the Kyiv Reservoir and, in particular, in its upper section, which accepts the major part of the inlet from the Pripyat River sediment load.

Relocation of contaminated areas in the water area of the reservoir has been observed in recent years. The most contaminated fine silt particles have been suspended and transported to the middle and lower parts of the Kyiv Reservoir, where these particles are re-deposited. On the other hand, natural attenuation processes are also taking place. The contaminated sediment is being covered with a layer of less contaminated material. In the next decade, sedimentation will play a dominant role in creating a relatively clean sediment layer above the buried radioactive contaminated particles in the deepest areas of the Kiev reservoir.

Radionuclides in the groundwater

Contamination levels in the groundwater remain relatively low, excluding those sites around temporary waste disposal sites and some radioactive storage facilities situated at the ChNPP industrial site. Groundwater is also significantly contaminated in areas adjacent to the contaminated water bodies (cooling ponds, lakes and wetlands in the most contaminated areas of the ChEZ). In the worst contaminated areas of the ChEZ, the level values vary in range from 10^2 and less to 10^3 Bq m^{-3} for ^{137}Cs and between 10^3 and 10^4 Bq m^{-3} for ^{90}Sr.

Both ^{137}Cs and ^{90}Sr contamination of the groundwater in the first aquifer under the surface registered beyond the Exclusion Zone boundaries does not exceed 0.3×10^3 Bq m^{-3}. In the water of Cenomanian and Lower

Cretaceous aquifer system, ^{90}Sr and ^{137}Cs volumetric activities have not exceeded $10\,Bq\,m^{-3}$.

11.4.2 Experience of territory decontamination

The need for protective actions became evident very soon after the Chernobyl accident occurred. Activities for area decontamination were part of an extensive set of short- and long-term environmental countermeasures, applied to protect workers and the public from radiation. These countermeasures involved large amounts of human, economic and scientific resources. According to IAEA (2001), such countermeasures included:

- reduction of radionuclides release from the destroyed reactor (in the early stages of the accident),
- evacuation of population and its resettlement,
- construction of the 'Shelter' object (SO),
- decontamination of the soil, buildings and installations,
- disposal of the RAW resulting from the decontamination measures,
- surface and groundwater protection,
- restriction of access to the contaminated areas and the prohibition of economic activity,
- changing the type of forestry and agricultural activities,
- ban or limitation of the consumption of contaminated foodstuffs,
- reduction of radioactive contamination of agricultural products,
- information to the population, social and other supplementary measures.

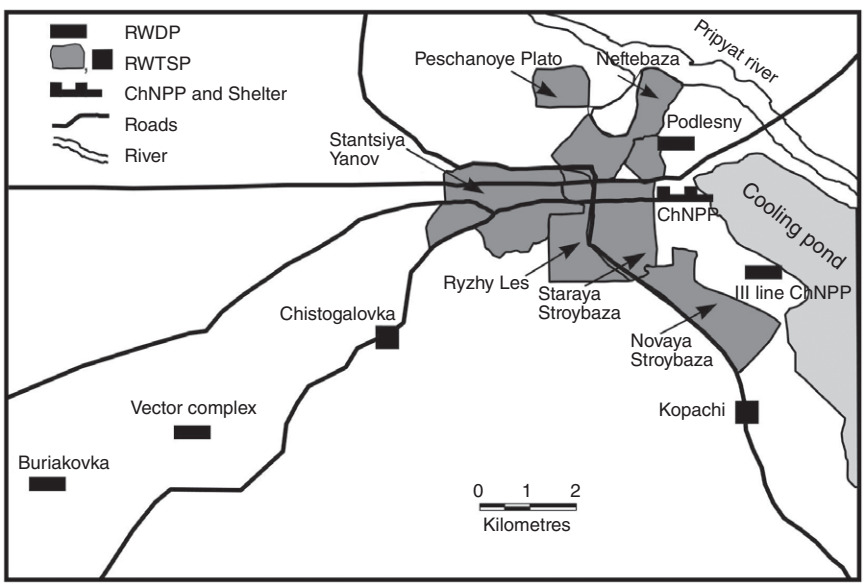
Large-scale decontamination and clean-up activities were performed between 1986 and 1989 both within the Chernobyl Exclusion Zone, and in the cities and villages of the USSR most contaminated after the Chernobyl accident. This activity was performed usually by military personnel and included removal and disposal of contaminated soil and civil constructions, cutting and disposal of contaminated forests, washing of buildings with water or special solutions, cleaning and washing of roads, and decontamination of open water supplies. The decontamination has produced a huge amount of radioactive waste, which was collected in numerous RWDP and RWTSP and has created a problem for its final disposal. More than 800 waste localization sites were created within ChEZ and 47 outside of it, with a total volume of more then $10^6\,m^3$. The reliability of these sites is a cause for concern, and the problem needs to be solved in the future. The greatest amount of RAW arose during decontamination of the site of the destroyed unit 4 in the course of the 'Shelter' object (SO) construction. The object is classified now as a temporary storage of radioactive waste (RSNU, 1997). The SO, RWDP and RDTSP are described in Section 11.4.3.

The efficiency of various measures for protecting workers and public from radiation was assessed in IAEA (2006). According to this source, depending on the decontamination technologies used, the dose rate was reduced by a factor of 1.5–15. However, the high cost of these activities hindered their comprehensive application to all contaminated areas. Due to these limitations, the actual effectiveness of the decrease in annual external dose was 10–20% for the average population and ranged from about 30% for children to less than 10% for outdoor workers.

It should be noted that in many cases the decisions about decontamination in 1986–1989 were of a political nature. In many cases such political decisions were in contradiction with conclusions reached by cost–benefit analysis. Therefore, the resulting effect in reducing the exposure dose was achieved by unnecessarily high costs in human and economic resources.

11.4.3 Current state of RAW management in the Chernobyl Exclusion Zone

Inside the Chernobyl Exclusion Zone, RAW management activities are carried out by the State Specialized Companies (SSC) of the Ministry of Emergency: SSC 'Chernobyl NPP', SSC 'Complex' and SSC 'Technocentre'. The locations in the Chernobyl Exclusion Zone where radioactive wastes are stored and buried is shown in Fig. 11.2.



11.2 Location of radioactive waste storage and disposal facilities within ChEZ.

RAW management at the ChNPP

SSC 'Chernobyl NPP' is carrying out the waste management activities at ChNPP and SO. Collection of ChNPP liquid waste is performed with a pipeline system. Accumulated liquid waste is stored in two tank stores at the ChNPP site. These storage facilities are a system of reservoirs made of corrosion-resistant steel, which are designed to accept 26,000 m^3 (liquid RAW storage facility) and 12,000 m^3 (liquid and solid RAW storage facility) of waste. This is the low and intermediate level waste: evaporation bottoms, pulp of spent ion exchange resin and pearlite pulp. Spent radioactive oil is also held in temporary storage tanks.

There are two storage facilities for solid waste at ChNPP site. The first storage facility is a surface concrete structure, which is divided into three groups of compartments for storage of LLW, ILW and HLW. The capacity of compartments is 1,087 m^3, 1,005 m^3 and 1,884 m^3 respectively. The second storage facility comprises 26 compartments for LLW, ILW and HLW. Its total volume is 10,000 m^3. Waste storage facilities are equipped with special protection systems.

Low and intermediate solid waste, generated as a result of work on termination of operation of power units and activities to transform the 'Shelter' object into an ecologically safe system, are collected and moved to the RWDP 'Buriakivka'. At the same time, high level solid waste is collected in the primary steel 200 L containers. Primary containers are inserted into shielding containers weighing about 4,000 kg, made of steel-reinforced concrete. Shielding containers are placed in a special temporary storage at the ChNPP site.

SSC ChNPP continues construction of facilities for RAW management with international financial support. These are the Liquid Radioactive Waste Treatment Plant and Industrial Complex for Solid Radioactive Waste Management (ICSRWM). ICSRWM includes:

- Lot 0 – Interim storage of low and intermediate level long-lived and HLW, which is constructed inside the ChNPP building.
- Lot 1 – Facility for removal of solid waste from their stores.
- Lot 2 – Solid waste processing plant.
- Lot 3 – Near-surface storage facility for solid waste at the site of the 'Vector' complex.

The 'Shelter' object

The SO (Fig. 11.3), which was intended to provide the environmental containment of the damaged reactor, was erected between May and November 1986, under conditions of high radiation exposure of the personnel. The SO was constructed using steel beams and plates as structural elements. Its

11.3 General view of the 'Shelter' object (as of 2008).

foundation rests at some points on the original structural elements of unit 4, whose structural integrity is not well known. Thus the ability of the SO structure to withstand natural events such as earthquakes and tornados is not known with any certainty.

The SO has approximately 1,000 m^2 of openings in its surface. These openings allow approximately 2,000 m^3 per year of precipitation to percolate through the radioactively contaminated debris. The collected water is contaminated with ^{137}Cs, ^{90}Sr and transuranic elements. The main potential hazard associated with the SO is a possible collapse of its top structures and the release of radioactive dust into the environment. Another concern related to the fuel-containing material is its possible transport out of the SO into groundwater through the accumulated water.

To avoid a collapse of the SO, some measures have been implemented to strengthen unstable parts of the SO and to extend their stability to 40 years. In addition, a new safe confinement (NSC) facility is planned to be built as a cover over the existing SO. The Ukrainian government supports the concept of a multifunctional facility with a service life of at least 100 years. This facility aims to reduce the probability of SO collapse, reduce the consequences of such collapse, improve nuclear safety, improve worker and

environmental safety, and convert unit 4 into an environmentally safe site. The construction of the NSC is expected to allow the current SO to be dismantled and removal of fuel-containing materials from unit 4.

RAW management at the SSC 'Complex' and SSC 'Technocentre'

SSC 'Complex' is responsible for the final disposal of waste at the RWDP 'Buriakovka', as well as for the monitoring of the RWDPs 'Podlesny', 'The III line of ChNPP', and multiple RWTSP (see Fig. 11.2). SSC 'Complex' also carries out the RAW collection and transportation within the ChEZ.

As the 'Buriakovka' facilities do not fully comply with the current requirements of surface disposal facilities, SSC 'Technocentre' started construction of the 'Vector' complex. At present, this new near-surface facility for low and intermediate level radioactive waste processing, storage and disposal, is under development. This complex will include:

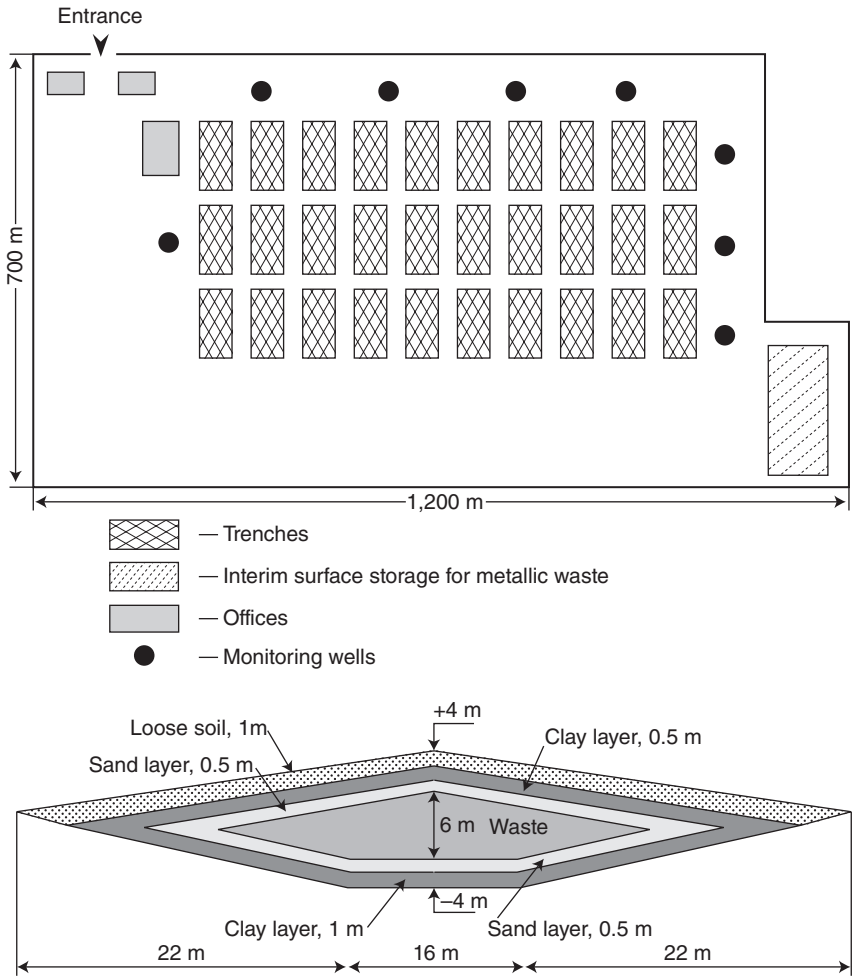
- an engineering facility for the processing of all types of solid RAW;
- disposal facilities for short-lived solid RAW;
- storage facilities for long-lived waste (including fuel-containing materials);
- intermediate storage for vitrified HLW to be prepared for final disposal at a deep geological disposal facility.[1]

RAW disposal points

The main radioactive waste disposal points for accident waste are the Buriakovka, Podlesny and III line of ChNPP sites. These three near-surface disposal sites were established after the accident to dispose of RAW from remediation actions carried out during the first year following the accident. These sites were chosen and designed for the disposal of higher level accident waste than the RAW located in the temporary RAW store points.

Buriakovka, built in 1987, is the only disposal facility currently in operation in the ChEZ. It comprises 30 trenches covered with a 1 m clay layer (see Fig. 11.4). Up to 590,000 m^3 of RAW has been disposed of, with a total

[1] In Ukraine, activities to create a deep geological repository have been carried out since 1993. They are performed by Institutes of the National Academy of Sciences of Ukraine and the enterprises of the State Geological Survey. This activity refers to the early stages of a siting and conceptual repository design. It is assumed that the most promising host rocks in which to locate a geological repository are Archaean and Proterozoic crystalline rocks of the Chernobyl Exclusion Zone and its vicinity. Two possible options for the repository design are considered: a mine (KBS-3 concept, Sweden) and a borehole one (VDH concept, Sweden). Further information can be found in Shestopalov *et al*. (2005, 2008).



11.4 General layout (upper figure) and separate trench cross-section (lower figure) of the 'Buriakovka' repository.

radioactivity of 2.5×10^{15} Bq of solid short-lived low and intermediate level waste. It consists of metal, soil, sand, concrete and wood contaminated with ^{90}Sr, ^{137}Cs, ^{134}Cs, ^{238,239,240}Pu, ^{154,155}Eu and ^{241}Am.

The Podlesny vault-type disposal facility was commissioned in December 1986 and closed in 1988. The facility was designed for the disposal of HLW with a dose rate 0.1 m from the surface in the range of 0.05–2.5 Gy h^{-1}. The total RAW volume of 11,000 m^3 of building material, metal debris, sand, soil, concrete and wood was placed in two vaults. The disposal facility was covered with concrete at its closure. In 1990 the estimated total radioactivity of the disposed waste was 2,600 TBq.

The III line of ChNPP vault-type facility was based on reconstructed facilities of the unfinished units 5 and 6 at the ChNPP site. This facility was in operation from October 1986 until 1988 and was designed for low and intermediate level waste corresponding to dose rates up to 0.01 Gy h^{-1} at 0.1 m from the surface of the waste container. More than 26,200 m^3 of solid waste with a total activity of 4×10^{14} Bq was disposed of in 18,000 containers and later covered with sand and clay. This waste is mainly sand, concrete, metal, construction material and bricks. Due to the high level of groundwater at different periods of the year, the facility is flooded 0.5–0.7 m above its bottom.

RAW temporary storage points

In the course of remediation activities in the vicinity of the ChNPP site, large volumes of RAW were generated and placed in temporary near-surface storage facilities located at distances of 0.5–15 km from the ChNPP site. The RWTSP in the shape of the trenches and landfills were created from 1986 to 1987 as a result of the clean-up of contaminated areas, to avoid spreading of dust, to reduce radiation levels and provide better working conditions at unit 4 and its surroundings. These facilities were established without proper design documentation, engineered barriers or hydrogeological investigations, which are required by contemporary waste safety standards.

The total area of temporary RAW facilities is about 8 km^2, with the total volume of disposed RAW estimated to be over 10^6 m^3. The main inventories of activity are concentrated in the Stroybaza and Ryzhy Les RWTSP along the western trace of the Chernobyl fallout. The specific activity of the RAW in the RWTSP at Ryzhy Les is 10^5–10^6 Bq kg^{-1} of ^{90}Sr and ^{137}Cs and 10^3–10^4 Bq kg^{-1} of plutonium isotopes.

Most of the facilities were built in the form of trenches 1.5–2.5 m deep in the local alluvial sand. The radioactive material (soil, litter, wood and building debris) is overlain by a layer of sand 0.2–0.5 m thick. These facilities are therefore very variable with regard to their potential for release, the retention capacity of the substratum along migration pathways and the location of the sites in hydrogeological settings. Only half of these temporary RAW facilities have been studied.

11.5 Problems and lessons learned

A comprehensive analysis of the status and existing problems in the field of RAW management in Ukraine was carried out in 2006–2007 within the framework of the TACIS project – U4.03/04: Development of the National Strategy and Concept for State Programme for Radioactive Waste

Management in Ukraine, including a Strategy for National Company Energoatom Radioactive Waste Management. The results of the project implementation were published in Shestopalov *et al*. (2008) and the following conclusions regarding radioactive waste management in Ukraine were made (as of 2008).

Ukraine has accumulated significant amounts of RAW. The volume of waste and the rate of its accumulation will continue to grow in the future due to the extension of the operating period and decommissioning of the existing units and commissioning of new NPP units. The contribution of NPP to the current accumulation of RAW in Ukraine is about 95%. In general, wastes have not been sorted or reprocessed taking into account the need for further conditioning and disposal. Separation of waste into short-lived and long-lived is not carried out at NPPs.

Unprocessed RAW from the non-nuclear sector continues to accumulate without being buried. Safety of the already buried waste has not been confirmed. The issues of storage and disposal of vitrified HLW as a result of reprocessing in the Russian Federation of Ukrainian NPP SF and long-lived waste of Chernobyl origin have not been addressed. The existing system of RAW management is not focused on the final disposal of all types and categories of RAW. The organization responsible for developing and implementing the technical policy in the field of RAW disposal has not been identified.

Stable funding of the design, construction and operation of infrastructure facilities for RAW management has not been ensured, and a special state fund for RAW management has not been established. The existing classification of RAW in Ukraine ensures the safety of RAW management at the stages of their collection and storage, but is economically inefficient in terms of achieving its ultimate objective the safe disposal of RAW. This is particularly true regarding the problems of disposal of the large amounts of waste of Chernobyl origin, which contain significant amounts of long-lived radionuclides.

The amount of funding for RAW management provides the minimum acceptable level of safety. State investment in RAW management infrastructure upgrades is virtually zero and a reassessment of the safety of storage facilities has not been made. Production of containers for the storage of RAW has only commenced in recent years. However, transportation of waste has not yet been provided by licensed transport containers.

Most of the problems of RAW management in Ukraine arose due to the lack of a single state policy that would allow a systematic solution to the problems of RAW management (including disposal) and the problems of stable funding.

11.6 Future trends

In recent years, Ukraine has made considerable efforts to resolve the above-mentioned problems. The main results of these efforts are:

- adoption by Parliament in 2008 of the law 'On State Purposeful Ecological Program on Radioactive Waste Management' (see State Program, 2008);
- development and adoption in 2009 of a Resolution of the Cabinet of Ministers regarding the Strategy for Radioactive Waste Management (see Strategy, 2009);
- adoption in 2009 of a number of alterations to the laws of Ukraine and new Resolutions of Cabinet of Ministers, which clearly defined the mechanism for accumulating and the procedure for spending the State Fund of Radioactive Waste Management (see State Fund, 2008);
- initiating the process of forming a national organization for RAW management.

The *State Program* (2008) defines the technical policy in the field of RAW management for 2008–2017. Key tasks of this program are:

- construction of the 'Vector' complex (surface repository for all flows of short-lived waste);
- improvement of NPPs' RAW management;
- development of a facility for interim storage of HLW, which will be returned to Ukraine, after reprocessing of Ukrainian spent fuel in the Russian Federation;
- improvement of RAW management, which arose as a result of the Chernobyl accident;
- conversion of SISP of UkrSA 'Radon' for collection and container storage of RAW;
- improvement of state system of RAW accounting;
- development of RAW management infrastructure at Chernobyl NPP and at the 'Shelter' object;
- activities on site selection for RAW geological disposal facility.

The *Strategy* (2009) developed is to cover the next 50 years. The main principle of the Strategy is the conformity of all directions of activity to the ultimate goal of RAW management, namely, the safe disposal of wastes in central repositories. The tasks and measures of the first stage of the Strategy to 2017 are identified in the State Program.

The Strategy and the State Program provide two approaches for RAW treatment and conditioning:

- at sites of enterprises whose activity generates large volumes of RAW (e.g., Ukrainian NPPs);
- at centralized regional waste management facilities designed for enterprises which use small volumes of radioactive materials (e.g., enterprises in the non-nuclear sector).

RAW has to be disposed of in centralized disposal facilities. To construct and operate these facilities requires development of the infrastructure for RAW management. To dispose of long-lived and high-level RAW, the Strategy provides for construction of disposal facilities in deep geological formations. A complex of centralized disposal facilities intended for disposal and long-term storage of RAW is under construction at the 'Vector' site in the Chernobyl Exclusion Zone.

The *State Fund* is intended to solve tasks and measures of the State Program. The mechanism for accumulating costs for the State Fund is based on the 'who contaminates must pay' principle. In accordance with the adopted regulations, all enterprises and organizations in the territory of Ukraine whose activities result or may result in radioactive waste must pay fees to the Fund and receive guarantees that the State will ensure further safe management of the RAW generated, including its final permanent disposal.

In 2011 the Ministry of Emergency Situations of Ukraine started the process of establishing the National Authority of Public Administration and the National Organization for Radioactive Waste Management. According to the existing plans, the formation of a unified technical policy, the administration of RAW management and management of spending the state funds will be carried out by the National Agency for Management of the Chernobyl Exclusion Zone. On the basis of the existing enterprises (SSC 'Complex', SSC 'Technocentre' and UkrSA 'Radon'), a single centralized state specialized enterprise, which will be responsible for the establishment and operation of centralized disposal facilities, is being created.

11.7 Conclusion

Significant amounts of RAW were accumulated during the period of nuclear power use in Ukraine. The rate of their accumulation will increase in future due to plans for further development of nuclear power. Additionally, large amounts of waste arose as a result of efforts to limit the consequences of the Chernobyl accident.

The system and infrastructure for waste management inherited from the Soviet Union has not allowed the existing or emerging problems to be solved. Since the collapse of the Soviet Union, much attention has been

paid in Ukraine to legal, organizational, financial and technical measures to improve the system of RAW management.

The Ukrainian system of RAW management is developing dynamically. Directions for development are determined on the basis of analysing its own experience, and learning from advanced international know-how. The development is financed by the government of Ukraine and within the framework of international technical assistance programmes. In the short term, creating the conditions for sustainable development of nuclear power in Ukraine depends on success in solving the current problems of RAW management and, in particular, on progress in creating a mechanism for permanent waste disposal.

11.8 Sources of further information

Sources of additional information on various aspects of radioactive waste management in Ukraine are national reports and annual regulator's reports.

Two types of national reports are periodically issued in Ukraine:

- On Compliance with the Obligations under the Joint Convention on the Safety of Spent Fuel Management and on the Safety of Radioactive Waste Management (Joint Convention Reports);
- Reports on the experience of eliminating the consequences of the Chernobyl accident (Chernobyl reports).

Joint Convention Reports are issued every three years by the State Nuclear Regulatory Inspectorate. These reports contain official information on the list of nuclear facilities, the amount and properties of RAW, technical policy in the field of RAW management, and current and planned waste management practices in Ukraine (e.g., NatRep, 2008). These reports are translated into English. They can be found on the website of the State Nuclear Regulatory Inspectorate: http://www.snrc.gov.ua/nuclear/en/publish/article/119836.

Chernobyl reports are published every five years by the Ministry of Ukraine of Emergencies. These reports contain official information about the cause of the accident at the Chernobyl NPP and the experience of attempts to limit its consequences, the current state of environmental contamination, state of the 'Shelter' object and its transformation into an ecologically safe system, as well as on radioactive waste management in the Chernobyl Exclusion Zone (e.g., ChernNatRep, 2011). These reports are translated into English and are published in the form of books.

Annual Regulator's Reports are published every year by the State Nuclear Regulatory Inspectorate. These reports contain concise official information on the list of nuclear facilities, RAW volumes, the current practice of waste management and irradiation doses of the personnel and

the population (see SNRCU, 2009). These reports are translated into English. They can be found on the website of the State Nuclear Regulatory Inspectorate: http://www.snrc.gov.ua/nuclear/en/publish/article/119456.

Additional information in English about RAW management in Ukraine, waste of Chernobyl origin and the experience in eliminating the consequences of the accident can also be found on the websites of the IAEA and the European Commission.

11.9 References

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12

Czech Republic, Slovak Republic and Poland: experience of radioactive waste (RAW) management and contaminated site clean-up

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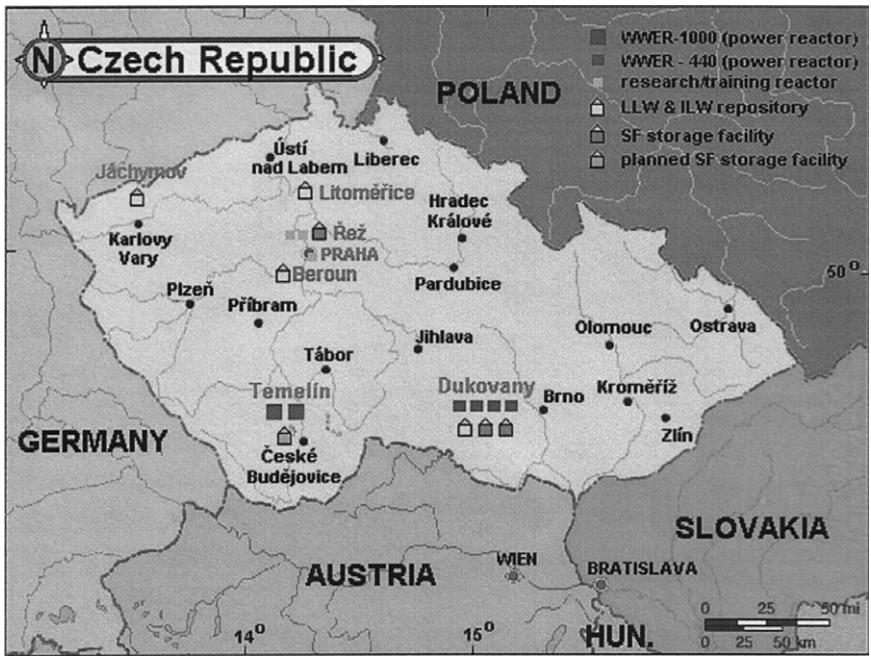
Abstract: The chapter describes radioactive waste (RAW) issues in the Czech Republic, Slovak Republic and Poland. The situation in the Czech and Slovak Republics is different from Poland. Poland has run only experimental reactors, while the Czech and Slovak Republic have operated nuclear power plants (NPPs) since the 1970s. The Czech and Slovak nuclear programmes were based on the assumption of returning reactor spent fuel (SF) assemblies to the Soviet Union without any commitment concerning SF destiny. After the decision of the Russian Federation to cancel 'free of charge' returning to the Russian Federation, both countries started to develop their own concept concerning SF disposal. The main problem for Poland is that their repository at Różan for RAW from industry, medicine and research is almost full and it necessitates finding a new facility for accepting waste by 2020.

Key words: radioactive waste (RAW), Czech Republic, Slovakia, Poland, spent fuel disposal.

12.1 Introduction

This chapter is devoted to the description of the radioactive waste (RAW) management situation in the Czech Republic, Slovak Republic and Poland. The situation in the Czech and Slovak Republics is different from Poland: Poland has not yet run any nuclear power plant (NPP), and is only considering starting its first NPP by 2020, whereas both the Czech and Slovak Republics have operated NPPs since the 1970s. Poland does, however, operate a number of experimental nuclear reactors. Figure 12.1 shows the locations of nuclear installation in the Czech Republic.

All of these countries are considering building new NPPs. Today in the region, public acceptance of nuclear energy is quite high (generally about 60%, in some countries up to 70%). Nevertheless, because of the possible consequences far beyond national borders in case of an accident, and because nuclear energy is also such a divisive issue among various



12.1 Locations of nuclear installations in the Czech Republic.

opponents, there is no doubt that, for each project to build new NPPs will require full and frank information regarding three key issues to be set out (IAEA, 2009):

- nuclear safety,
- non-proliferation, and
- RAW and spent fuel (SF) management.

Lessons learned from building and operating RAW and SF management systems in these countries, as described in this chapter, may, therefore, significantly contribute to preparing new, improved systems of RAW and SF management already included in the designs of the new NPPs.

In the case of the Czech Republic, this chapter will focus mainly on summarising information from RAW management at two NPP with Russian WWER 400 and WWER 1000 reactors located at Dukovany and Temelin. Less attention will be devoted to RAW management from the use of ionising radiation in industry, medicine or research including SF management from research reactors. This is because, compared with waste from NPPs, waste from other sources is not so great a problem in the Czech Republic and management of this sort of waste was established in the 1960s and suitable disposal facilities are available. However, problems with remediation

of contaminated sites after extensive uranium mining and milling in the Czech Republic will be highlighted.

Decommissioning of nuclear facilities, in addition to RAW management at Jaslovske Bohunice and Mochovce NPPs with WWER 440 reactors, is a big issue in Slovakia, because of an operational incident at the first Czechoslovak NPP (A1) in 1977, after which it was shut down. In addition, the closure of the first generation of WWER reactors (V-230 type) at NPP (V1) at Jaslovske Bohunice was one of the conditions for fulfilling the Accession Agreement of Slovakia to the European Union.

The major topic for Poland concerning RAW management is primarily disposal of SF assemblies from research reactors at Świerk and management of radioactive waste from industry, medicine or research.

12.2 Sources, types and classification of wastes

12.2.1 Czech Republic

According to decree 307/2002 Coll. of Czech Regulatory Body (State Office for Nuclear Safety (SONS)) radioactive waste in the Czech Republic is classified as gas, liquid and solid. Solid RAW is classified into three basic categories: temporary, low- and intermediate-level, and high-level wastes. Temporary waste is waste whose radioactivity after long-term storage (maximum 5 years) does not exceed the clearance levels. High-level radioactive waste is waste for which heat generation must be taken into account during its storage and disposal. Other radioactive waste is classified as low- and intermediate-level waste. Low- and intermediate-level waste is classified into two subcategories. The first subcategory is short-lived waste, in which the half-life of radionuclides contained is shorter than 30.2 years with a limited mass activity of long-lived alpha emitters (in individual packages a maximum of 4,000 kBq/kg), and a mean value of 400 kBq/kg in the total volume of waste produced in a calendar year. The low- and intermediate-level waste can be accepted into near-surface repositories. The limits of radionuclides in the disposal facilities are calculated on the basis of safety assessments for individual storage and disposal facilities. The standard document must be issued for all types of RAW and for each RAW package that is an independent manipulation unit. This standard accompanying document contains a number of parameters, such as physical and chemical form or mass activity of radionuclides whose content will be limited by acceptance criteria.

Spent fuel assemblies are not considered as RAW unless they have been classified as such by the owner or by the SONS. Natural materials produced in the course of mining and treatment of uranium ores are not considered as RAW.

From the technical point of view, RAW from NPPs is divided into a large number of categories, such as: waste water, sludge, ion-exchange resins, sorbents, oils and solvent, solid compactable and combustible waste (personal protective equipment, decontamination and cleaning equipment, packaging materials, paper, polymer materials), non-combustible waste (glass, wires, cans, metal materials, ceramics, filters), wood, and large metal objects.

Another interesting RAW categorisation is related to the management of SF, and selected categories of RAW are shown in Table 12.1 (Czech National Report, 2008). The largest amount of RAW in the Czech Republic comes from NPPs. The Czech Republic operates 4 WWER 440 reactors at Dukovany and 2 WWER 1000 reactors at Temelin. The four 440 MW Dukovany units were installed and started operation during the period of 1985–1988. The two WWER 1000 reactors at Temelin started operation in 2002 and 2003.

Spent fuel

Currently, SF is stored in dry storage facilities located in the area of both NPPs in CASTOR-440/84-type approved casks or in pools at reactor sites. More than 9,000 SF assemblies from WWER 440 reactors and 1,000 spent assemblies from WWER 1000 reactors are stored in this way. More than 5,300 assemblies are expected to be spent by 2025 at Dukovany reactors and 2,600 assemblies by 2042 at Temelin reactors.

The multi-billion euro contract to build two new nuclear reactors at the current site of Temelin with the option for another three elsewhere has been launched recently in the Czech Republic; one in Dukovany and the second in Slovakia. The reactors will likely be built by US or Russian companies. It is expected that more than 8,000 fuel assemblies would be spent in the three new nuclear reactors in the Czech Republic during their 60 years of electricity production.

Other SF assemblies are from the research reactor located at the Nuclear Research Institute (NRI) Rez located near Prague. This experimental nuclear reactor has been in operation since 1957, with significant reconstruction performed in 1988–1989. Several hundred SF assemblies have been produced during this time. In 2004 the Czech Republic was included in the Global Threat Reduction Initiative (GTRI) programme. Highly enriched uranium (HEU) SF was shipped to the Russian Federation for reprocessing with the financial support of the US government and Department of Energy. To date, 457 spent fuel of IRT-2M and 208 EK 10 assemblies from the NRI research reactor have been sent to Russia. It is expected that after 20 years the corresponding activity will be returned back to the Czech Republic in the form of vitrified waste. NRI now also

Table 12.1 Categorisation of radioactive waste in the Czech Republic according to waste management

Type of liability	Long-term management policy	Current practice/facilities
Spent fuel	Preferred alternative – direct disposal in deep geological repository (DGR), but other alternatives are not excluded (reprocessing regional repository)	Long-term storage
Nuclear fuel cycle waste	Disposal in operating repositories and in planned DGR	Disposal in the operating repostory at Dukovany and storage in operating systems (NPPs)
Institutional waste	Disposal in operating repositories and in planned DGR	Storage and disposal in operating repositories (Richard, Bratrství, Dukovany) and storage (NRI Rez)
Decommissioning waste	Deferred dismantling (NPP) and immediate dismantling (research reactors)	Periodical review of decommissioning plans; all nuclear installations (NPPs, research reactors, storage facilities) are currently in operation
Disused sealed sources	Disposal in operating repositories and in planned DGR; return to the country of origin	Storage and disposal in operating repositories
Mining and milling waste	Tailing pond rehabilitation	Recovery of chemical uranium production on the Stráž site and use of tailing ponds on the Rožná site

participates in shipment of spent fuel assemblies from other Eastern European countries' research reactors to Russia, 'Mayk' Production Association. These reactors are of Russian provenance.

Operational waste from NPPs

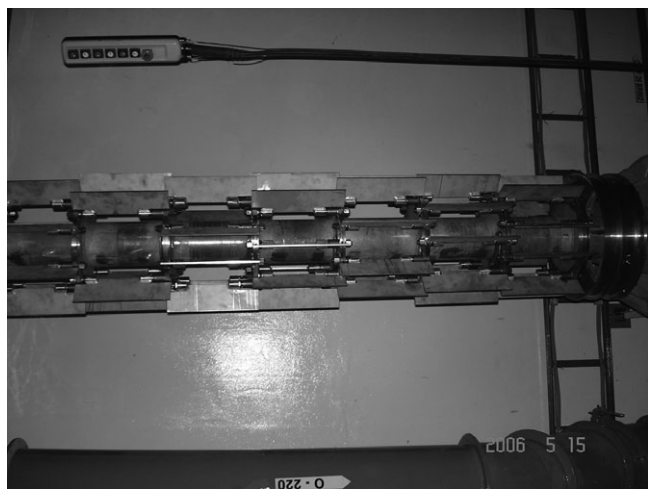
The original Soviet design concept for the waste management of Dukovany WWER reactors envisaged the collection, pre-treatment and interim storage of all radioactive waste produced at the site during the whole NPP operational period. Such a concept postponed the final decisions on

conditioning and disposal of operational waste to the decommissioning stage. Dukovany was therefore equipped with huge liquid storage capacity: 5,320 m^3 for concentrates and 920 m^3 for spent ion-exchange resins. A very similar concept was designed at Temelin, but with smaller storage capacity: 520 m^3 for concentrates and 200 m^3 for spent ion-exchange resins.

The disadvantage of the system of wastewater treatment in Dukovany NPPs, and partly also in Temelin NPP, is mixing of all sorts of wastewater, which complicates recycling of separate wastewater streams. Wastewater is routed to drain sump tanks from which it is pumped to a sedimentation tank and then through an overflow tank to wastewater holding tanks. Wastewater from laundry and laboratories is collected in control tanks and if it is not possible to discharge it, it is pumped to the radioactive drain sump tanks. Wastewater in holding tanks usually contains 0.5–2 g of salt/l with pH ~ 8 (Kulovany, 2001).

Temelin NPP adopted design changes, which allowed the reduction of the volume of wastes produced, dividing the draining system for multiple independent systems, so that inactive water can be discarded out of the primary circuit.

In both NPPs, evaporation is used to increase the salt concentration in the waste liquid by ~50. The basic equipment consists of a layer rotor evaporator with a vertical double-shell drum (rotor part of evaporator, produced by Czech company VUCHZ, a.s., is shown in Fig. 12.2). Prior to concentration, it is necessary to increase the alkalinity of the wastewater by addition



12.2 Rotor part of a layer rotor evaporator used for increasing the salt concentration in the waste liquid and bituminisation of liquid radioactive waste.

of sodium hydroxide to pH at least 11 to reach solubility at least 60 g of boric acid/l. The evaporator bottom contains a great amount of nitrates coming mainly from the recovery of cation-exchange resins by nitric acid. The problem facing the operators is to keep the boric acid in a soluble state to prevent its crystallisation. The evaporator bottom concentrates are solidified by bituminisation. The asphalt and liquid concentrate are tangentially sprayed to the upper part of the evaporator. Drained bitumen product, containing 30–40% of salts, flows down the evaporator wall and through heated piping to drums, which are transported to the near-surface, fully engineered disposal facility at Dukovany (Fig. 12.3). A special thermal resistance test of bitumen compound with evaporator bottom has to be carried out before bituminisation to prevent fire hazard. For this reason, the limit for manganese content in bituminised salt is 15 g/l and for pH 11.5.

Most ion-exchange resins are currently stored in storage tanks, but the State Office for Nuclear Safety (SONS) has recently approved their conditioning using geopolymers developed in the Slovak Republic (Majersky *et al*., 2007) and the first drums (20 m^3 of 307 m^3 stored) were already disposed of in the near-surface repository at Dukovany.

Liquid radioactive wastes of organic origin (oils) are stored in 200 l metallic drums. Currently, approximately 1.3 m^3 of this type of waste, are stored.

The solid waste bags are collected before treatment and sorted for detailed segregation of active and nonactive wastes. The activity of bags is measured using the system Merlin (Envinet, a.s.) equipped with three Canberra HPGe Big Mac detectors. The Merlin system and procedure for releasing the waste to the environment or landfill sites from nuclear facilities must always be approved by the Czech regulatory body.

12.3 Fully engineered repository at Dukovany.

The only available technology for solid waste treatment is low-pressure compaction, waste crusher and cable insulation ripper. High-pressure compaction was used to minimise the final volume of solid waste in 1996 (using a rented high-pressure compactor). Intermediate-level waste is only fragmented (if practicable) and stored under control in the storage facility for radioactive items. Some part of the organic solid waste has recently been incinerated in Studsvik and only the ash will be disposed of at the Dukovany repository. The average volume of RAW from operation of Czech NPPs Dukovany and Temelin is given in Table 12.2.

Institutional waste

RAW from the use of radioactive materials in industry, medicine and research (institutional waste) has been disposed of, or stored, in three near-surface repositories, Richard, Bratrství and Hostim since the early 1960s (Figs. 12.4 and 12.5). The inventory of the abandoned limestone mine repository, called Richard, is given in Table 12.3 (Czech National Report, 2008).

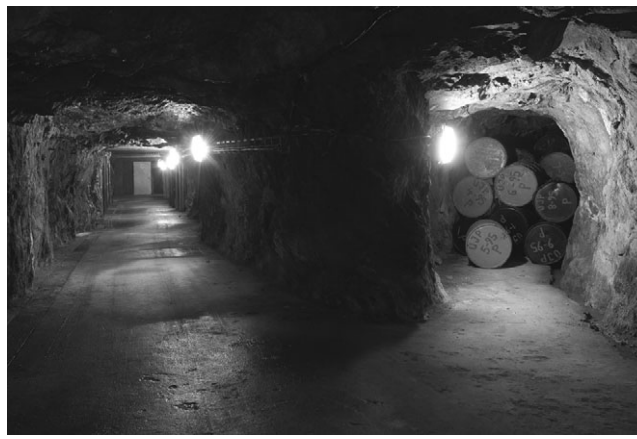
In addition to the waste disposed of, RAW that does not meet waste acceptance criteria and is awaiting disposal in a deep geological repository

Table 12.2 Operational waste from Czech NPPs

Type of waste	Dukovany NPP	Temelin NPP
Spent ion exchange resins and sludge	5–10 m^3/a	No spent ion-exchange resins have been produced so far
Evaporator concentrates	350 m^3/a	250 m^3/a
Dry solid waste	50 t/a	50 t/a

12.4 Drums with radioactive waste disposed of at Richard repository.

(DGR), described further in Section 12.3.1, is stored here. It consists mainly of sealed radionuclide sources of ^{137}Cs, ^{60}Co, ^{241}Am and ^{239}Pu. Inventory of the abandoned uranium mine Bratství located near the town of Jachymov, which has been in operation since 1974, is given in Table 12.4. The first near-surface repository, Hostim, near the town of Beroun, was closed in 1965. It contains only very low-activity waste not exceeding 10^{11}Bq.



12.5 Drums with radioactive waste disposed of at Bratrství repository.

Table 12.3 Inventory of radioactive waste in the Richard repository

Radionuclide	Total activity (Bq)
^{3}H	4.54×10^{13}
^{14}C	8.20×10^{12}
^{36}Cl	8.90×10^{9}
^{90}Sr	2.58×10^{13}
^{99}Tc	8.35×10^{7}
^{129}I	4.94×10^{6}
^{137}Cs	5.05×10^{14}
Total activity of long-term α radionuclides	1.52×10^{13}

Table 12.4 Inventory of Bratrství repository

Radionuclide	Total activity (Bq)
^{226}Ra	1.33×10^{12}
U	4.11×10^{11}
^{232}Th	1.34×10^{8}
^{90}Sr	2.58×10^{13}

12.2.2 Slovakia

The RAW categorisation and classification system of the Slovak Republic does not differ significantly from the system already described for the Czech Republic, because it was largely formed before Czechoslovakia was split into two countries in 1992.

The Slovak Republic now operates 4 WWER 440 reactors, two in Jaslovské Bohunice and two in Mochovce. Three reactors are now under decommissioning and two reactors of WWER 440 type are under construction.

The first Czechoslovak, gas-cooled, heavy water moderated reactor, located in Jaslovské Bohunice, known as A1, started operation in 1972, but was shut down in 1977 following an operational incident. The commitment to shut down two units of the first generation, Russian, WWER 440 reactors was taken in Slovakia in 1999 as a condition for fulfilling the Accession Agreement of Slovakia to the European Union. The waste from decommissioning is of various categories; most of the waste is low level, which can be disposed of at the near-surface repository at Mochovce (Fig. 12.6), but some of the waste, especially inorganic SF coolant, which contains almost 10% of the SF activity due to cladding failure, is vitrified in the pilot plant vitrification facility and will have to be stored until the availability of a DGR described further in Section 12.3.2.

Spent fuel

While SF assemblies for A1 NPP and a small number of assemblies from the V1 NPP were returned to Russia, more than 25,000 SF assemblies are

12.6 The Mochovce repository.

planned to be disposed of in a DGR. Some of them are now stored in the wet storage facility in Jaslovské Bohunice.

Operational waste from NPPs

Similar waste management concepts as described above for the Czech Republic WWER reactors are also used in the Slovak Republic. The Jaslovské Bohunice waste collection and storage systems were designed for 10 years of waste production for the double WWER 440 unit with 3,200 m^3 tanks for concentrates, 2,000 m^3 for spent ion-exchange resins and 5,000 m^3 of vaults for low-pressure compacted solid wastes (Burclová *et al.*, 2001). Due to a plant waste minimisation progamme implemented in 1999 (IAEA, 2006), the average amount of low- and intermediate-level waste from reactor operations has decreased significantly from the several hundred m^3 of liquid waste at one NPP (600 m^3/a) and 350 m^3/a of solid waste before 1999 to slightly more than 100 m^3 of liquid and solid waste at this time (IAEA, 2006). This programme continues to this day. According to Noferi (2009), it aims to reduce the amount of conditioned liquid RAW by 99% using advanced technologies, such as ozone oxidation of the complexes, the use of special sorbents or ultrafiltration.

A very significant effort was devoted to introducing new treatment and conditioning technologies. Bohunice's new waste treatment and conditioning centre, designed by NUKEM, was completed in 2001. In this facility, the solid and liquid burnable waste can be incinerated with two-stage burning with a throughput of 50 kg/h. A high-force compactor with compaction force 20,000 kN and throughput 10 drums/h is used for the volume reduction of solid waste. A cementation plant is used for the conditioning of liquid concentrates, incinerator scrubber saturated liquids, and encapsulation of non-compactable dry waste placed in drums. Cement is also used for the final filling of void spaces in disposal containers that are pre-loaded in drums with bituminised waste.

All waste from reactor operation is disposed of in special fibre-reinforced concrete containers (licence of French company Sogefibre) at the fully engineered near-surface repository at Mochovce with capacity of 22,320 m^3. Similar to the Czech Republic, however, the current inventory in this repository is low, because only solid waste and bitumenised evaporator bottom waste is disposed of at the Mochovce repository.

Institutional waste

Currently, about 3,000 sealed sources are present in Slovakia, 1,200 of which are no longer used and are stored at the user premises. The institutional waste is treated in the same way as operational waste from NPPs and,

after meeting waste acceptance criteria, disposed of at the Mochovce near-surface repository.

12.2.3 Poland

According to Polish Atomic Law, RAW is classified according to its activity level or exposure measured at the surface as low-, intermediate- or high-level waste. These categories can be divided into subcategories taking into account their half-life or generated thermal power. Another category is used sealed radioactive sources which are divided into short- or long-lived low-, intermediate- or high-level.

High-level waste independent of the SF's activity is mainly high uranium content waste especially from spent nuclear fuel or waste remaining after its reprocessing. Producers of such waste were research reactors. The first of these, Ewa, is already closed, while the second, Maria, is still working in the Institute of Atomic Energy (IEA) – POLATOM in Świerk near Warsaw. In Poland, spent nuclear fuel or uranium ore is not reprocessed, so there are no further sources of high-level waste. All other waste, generated by industry, hospitals, scientific and educational institutions, are characterized as low or intermediate.

For treatment, the radioactive waste in Poland is divided into gaseous, liquid and solid waste. The solid waste is divided into compressible or non-compressible.

The first Polish research reactor (Ewa), which was a Russian tank-type, was shut down in 1995 after 35 years of operation. The second research reactor (Maria), which is a 30 MW pool-type reactor, has been in operation in the Institute of Atomic Energy – POLATOM since 1975. The Maria reactor was designed mainly for material testing. Between 1985 and 1992 the reactor was shut down and modernised. The reactor is planned to work until 2020 and then, after further modernisation, from 2020 to 2050. The Maria reactor is now one of the best research reactors in Europe. It has power higher than 15 MW and neutron flux higher than 1×10^{14} n/cm^2, and is mostly used to produce radioisotopes, for materials testing, activation analysis, etc. Initially, the Maria reactor was supplied with highly enriched uranium (HEU) fuel with enrichment level up to 80%. Since 2002, for non-proliferation reasons, a low enriched uranium (LEU) fuel has been used with enrichment level up to 36%.

Spent fuel

About 4,000 SF assemblies have accumulated in the IEA from operation of the two reactors, which are temporarily stored in ponds at the IEA. For the first operational period of the Ewa reactor (1958–1967), the LEU

EK-10-type assemblies were used. By the year 2009, all of the EK-10-type fuel (about 2,600 assemblies) were encapsulated in stainless steel capsules filled with helium gas. After encapsulation, it can be stored dry. From 1967 to 1995 the HEU fuel WWR-SM and WWR-M2-type fuel were used in the Ewa reactor and about 800 assemblies are stored in a water pool at IEA. About 350 assemblies of the HEU MR-6-type fuel and about 15 assemblies of MR-5-type fuel from the Maria reactor are stored in the water pools at Świerk. About 158 SF assemblies from the Maria reactor were encapsulated from 2003–2007. In 2009, the GTRI programme was initiated and in September the first part of the WWR-type SF from the Ewa reactor was shipped to the Russian Federation.

Operational waste from experimental reactors

The annual generation of low and intermediate liquid level waste from Polish research reactor operation ranges from 30 to 160 m^3. The liquid waste is subjected to an evaporation process or is purified by sorption. The evaporator bottom concentrates are solidified by bituminisation or cementation and then disposed of at the near-surface type central repository at Różan. Annual production of solid waste is in the range 5–20 m^3. Solid waste is compacted into carbon steel zinc-plated drums and then transported to the Różan repository.

About 90% of the liquid waste originated from the Maria reactor operation, while the rest comes from radioisotope production or after decontamination. Annual production of the solid waste from industry, hospitals or research activities is in the region of 15–40 m^3, spent sealed sources of about 1,000 pieces and smoke detectors of about 20,000 pieces. There is some waste after uranium mining activities which took place in Lower Silesia in the south–west of the country which ended in 1968. There are some 100 dumps of waste rock and ore totalling approximately 1.4×10^6 m^3.

Institutional waste

The institutional liquid waste is concentrated by evaporation or sorption. The concentrate is then solidified by bituminisation or cementation in drums. The solid waste is compacted into drums by a hydraulic press. Such immobilised waste is then transported to the Różan repository.

The Różan repository is located about 90 km from Warsaw. The site was originally a military fort built at the beginning of the twentieth century covering an area of 3.2 ha. The repository has been operational since 1961 and is the only repository in Poland. The annual average activity of the waste sent to the Różan repository is between 1 and 2 TBq. The total quantity of waste which is assigned for final disposal is about 45 m^3 per year

(about 70 tons). The final volume is about 80 m^3 (about 35 m^3 is bonding material, mainly concrete). Solidified waste is placed in drums of volume 0.05 or 0.2 m^3. The Różan repository is planned to operate until 2020. During 50 years of operation, about 3,300 m^3 of different kinds waste of total activity about 34 TBq have been collected there.

12.3 Radioactive waste (RAW) management strategies: history and developments

12.3.1 Czech Republic

The uranium ore chemical processing plant in Jáchymov began the industrial production of uranium pigments in 1853 (at that time Jáchymov was named Joachimsthal and belonged to Austria-Hungary). The insoluble residue from uranium leaching was enriched by radium ^{226}Ra. Over more than 40 years and before the discovery of radioactivity, this by current standards worthless long-lived RAW was accumulated in this uranium plant in a form suitable for the subsequent separation of radium, which significantly facilitated the separation and isolation of the first pure radium discovered in 1898 by M. and P. Curie and G. Bémont. Following the takeover of both the mining and the uranium factory by the Czechoslovak government, the annual production of the factory was of the order of several grams of radium and about 10 tons of uranium pigments (Vobecký, 1999).

However, significant RAW accumulation and management started after the establishment of NRI at Rez in 1955, and particularly after the start of operation of the NPPs: the first Czechoslovak NPP A1 in 1972 in Jaslovské Bohunice (now belonging to Slovakia), the second NPP V1 (again now in Slovakia at Jaslovské Bohunice) in 1979, and the first NPP on the territory of the Czech Republic at Dukovany in 1985.

The Czechoslovak nuclear programme was based on the assumption of returning reactor SF assemblies to the Soviet Union (to the fuel manufacturer) without any commitment concerning SF destiny, including financial costs. This was why the first Czechoslovak concept of RAW management approved by the Czech government in 1981 dealt only with waste from reactor operation (Marek, 1993). This concept reflected the fact that NPPs of Soviet origin were not equipped with a complete nuclear waste treatment system. Wastes were stored within the power plant area with only one type of treatment – concentration of liquid waste. The basic principle of the concept was that liquid wastes must be solidified and placed into near-surface repositories in steel containers. One repository was then built in the Czech Republic (in operation from 1995 at Dukovany) and one in the Slovak Republic (in operation from 1999 at Mochovce). It was also decided that the waste treatment systems would be based on domestic technologies.

On the basis of this concept, bituminisation facilities, described above, were built both in the Czech Republic at Dukovany NPP and in the Slovak Republic at Jaslovské Bohunice NPP.

Concerning institutional waste, the NRI at Rez handled all liquid RAW not only from the Institute itself but from all institutions in the Czech Republic. The Institute of Research, Production, and Application of Radioisotopes focused on collection and disposal of all solid wastes including sealed sources. The first repository to be operational was in the village of Hostim near to the town of Beroun. This repository was closed in 1965. From that time until now, two repositories have served for disposal of institutional waste, Richard near the town of Litomerice for wastes with artifical radionuclides, and Bratrství near the town of Jachymov for natural radionuclides.

The first concept for a new RAW management strategy, following the decision of the Russian Federation to cancel 'free of charge' return of SF to the Russian Federation, was based on the decision that contracts with Russia are not held mainly for political reasons. It was decided that the RAW system will be guaranteed by the state and will respect two main axioms:

1. The state undertakes responsibility for the safe and final disposal of RAW to protect every citizen.
2. The holder of 'the state licence' becomes the owner of the waste.

On the basis of this concept, the Atomic Law was prepared and approved by Parliament in 1997. According to this Law, the Radioactive Waste Repository Authority (RAWRA) was established in 1997, which is now responsible for RAW and SF disposal activities. The Czech Power Company (ČEZ) is responsible for storage of SF assemblies. The Nuclear Account, into which radioactive waste generators pay fees, was also created in agreement with the 'polluter pays' principle.

In 2002 the Czech government approved a new RAW management concept that determined the main basic tasks that have to be performed for safe disposal of waste and SF generated in the Czech Republic. One of the main points of the concept is to dispose of SF assemblies directly in a DGR within the territory of the Czech Republic. In 1991, it was supposed that operation of the repository would start by 2032. This date has now been put back to 2065. One of the reasons is that SF assemblies can be safely stored in dry storage facilities at the NPP sites. The time of storage is sufficient for possible extraction of fissile products from SF and possible decreasing of radiotoxicity of the radioactive waste via as yet unproven technologies such as partitioning and transmutation.

The DGR is planned to be located in granite host rock, because no other type of host rock in sufficient volume is available in the Czech Republic.

SF assemblies should be located in vertical or horizontal boreholes at approximately 500 m underground and surrounded by compacted clay, bentonite. The first reference design of a repository was prepared in 1999. An update of the reference design was prepared in 2011, but the basic concept of DGR is the same as proposed in 1999.

Due to problems with local acceptance of sites for the repository, a decision was made by the Czech Ministry of Industry and Trade in February 2004 to suspend all on-site characterisation work for five years. However, after these five years, the situation is not much better. People from suitable candidate sites selected on the basis of already existing geological information and non-destructive methods still oppose the presence of DGR on their sites, but they are now more willing to discuss the conditions for starting the siting process.

12.3.2 Slovakia

The history of RAW management in Slovakia also starts in the former Czechoslovakia. After the division of the former Czechoslovakia into two countries in 1992, there were almost no consequences for the management of NPP wastes, because the systems were developed and implemented separately within both Republics. Only the system for institutional RAW management had to be newly developed in Slovakia, because organisations responsible for it remained in the Czech Republic.

The absence of a waste management system for institutional waste led to the acceptance of the first Slovak concept for RAW management in 1994. The important decision of the concept was that all the RAW from institutions would be treated in the same way as waste from NPP operations. The Slovak government accepted the resolution that the reference strategy for SF assemblies is their direct disposal to a DGR, but the possibility to reprocess them in future was not excluded. Until the final decision regarding potential reprocessing is made, the SF assemblies would be stored. The first version of the Slovak Atomic Law was accepted in 1998 and updated in 2004. It includes the basic requirements for RAW management, covering also the establishment of a new agency for RAW management. Until that time, RAW was governed by the National Nuclear Fund according to Law No. 238/2006 Coll. for decommissioning of nuclear facilities and RAW management. The board of this Fund prepares strategies for decommissioning and RAW management and approves also financial means for decommissioning and other activities. RAW management activities are divided between producer (collection, sorting, pre-treatment and treatment) and waste management organisation JAVYS owned by the state that operates most of the storage, treatment, conditioning and disposal facilities.

The current concept of Slovakia for SF management is still based on direct disposal of SF assemblies in DGRs. However, unlike the Czech Republic, the SF assemblies are stored in the wet storage facility at Jaslovské Bohunice, whose licence expires in 2037. After 2037, SF assemblies will have to be transported to another facility or the licence of the storage facility will have to be prolonged on the basis of safety analyses.

The development of a DGR in the Slovak Republic started in 1996. Granitic and clay sites suitable for the location of a repository were found in the territory of Slovakia. But the search process was stopped in the 1990s for political and/or financial reasons. Currently, a new programme for development of a DGR is being considered.

Low- and intermediate-level waste programmes in the Slovak Republic are very ambitious and a number of new, advanced technologies and facilities described below have been commissioned in last few years (see Chapter 6 for more detail). Nevertheless, with new plans for construction and extension of the lifetime of NPPs, and for building new nuclear reactors, Slovakia is facing a number of challenges connected with the need for new capacities both for storage of SF assemblies and disposal of low- and intermediate-level waste.

One of the most important issues in the Slovak Republic is decommissioning of the first Czechoslovak NPP A1 after an operational incident in 1977 and decommissioning of the first generation of WWER reactors (V-230 type) at NPP (V1) at Jaslovské Bohunice, which was one of the conditions for fulfilling the Accession Agreement of Slovakia to the European Union. Decommissioning of nuclear facilities is closely connected with generation of large amounts of waste of all categories. In particular, RAW from NPP1 represents a specific problem when considering the need to address the high-activity wastes and wastes containing long-lived, alpha radionuclides. The process of decommissioning is very slow and it is expected that it will not be finished earlier than 2033. A number of technologies were developed and tested in the first decommissioning phase. Bituminisation and pilot plant vitrification facilities were commissioned: the first in 1984 for bituminisation of A1 concentrates and the second for vitrification of SF coolant, Dowtherm in 1996. Special technology for solidification of high-activity sludge using geopolymers was developed (Majersky *et al.*, 2007). The Bohunice waste treatment and conditioning centre contains a variety of treatment and conditioning technologies (incineration, over-concentration, cementation, high-pressure compaction, bituminisation, fragmentation).

The second centre for the treatment and conditioning of RAW in Slovakia was established in the area of the NPP in Mochovce. This centre was designed primarily for the treatment of liquid RAW originated from NPP Mochovce. It is operating on a campaign schedule. Unlike in the Czech

Republic, spent resins and sludge are bituminised in a discontinuous batch-type facility (Hanusik *et al.*, 2008). This facility consists of a decanter, dryer and mixer-homogeniser. The sediments are isolated in the decanter and consequently flow by gravity into the dryer, heated by steam. The dried sediment of spent resins and sludge is then mixed with melted bitumen and discharged to 200 l drums. The cement grout is prepared by mixing evaporator bottom concentrate (400–450 g/l) with lime and a zeolite-cement mixture. High-integrity fibre-reinforced concrete (FRC) containers (internal volume of 3.1 m^3) are loaded with bitumen product drums and free voids filled with cement grout. These containers are then transported to the Mochovce repository (Fig. 12.6).

12.3.3 Poland

Polish nuclear waste management history started in 1958 when the first research reactor Ewa started operation in the former Institute for Nuclear Research (IBJ). At the beginning of the 1960s, rapid development of nuclear techniques in many industrial domains took place. There were no specific laws concerning managing, storing and processing of the nuclear waste at this time, and so there was a dangerous build-up of unmonitored and uncontrolled radioactive sources. To address the RAW problem, the Central Radioactive Waste Repository in Różan was opened in 1961, which is still accepting waste to this day. Initially, the Radioactive Waste Center (COP) in the former Institute for Nuclear Research was established. At this time the COP initiated technologies of nuclear waste management and reprocessing such as compressing, solidification and different techniques for immobilisation of radioisotopes. In 1970, the COP was transformed into the Radioactive Substance Management Plant, which became part of the Institute of Atomic Energy in 1983. In 2002 the Institute of Atomic Energy was transformed into the Radioactive Waste Management Plant – the state-owned Company (ZUOP). Since that time, it has been operated as a state-owned company of public utility and is the only institution in Poland that deals with management of RAW.

In 1982 Poland started to build a NPP in Żarnowiec in the northern part of the country. Four WWER 440-type reactors were planned. Changes in economic conditions and protests by ecological organisations caused the construction to be stopped. In 2009 the government voted for the Energy Policy of Poland until 2030 in which it is planned to build two new NPP of 3,000 MWe each and the first should start operation by 2020. At this moment, Poland is not going to locate a deep geological repository at least until 2050. The present radioactive waste repository at Różan is almost full, so it is necessary to find a location and prepare for a new facility which should start accepting low- and intermediate-level waste by 2020.

12.4 Contaminated site clean-up experience

Uranium mining and milling in many Eastern European countries caused enormous devastation of the environment due to the accumulation of waste at dumps left after uranium mining or tailings generation. The situation is most serious in the Czech Republic, because this country was the first source of uranium for the Soviet Union's atom bombs. Table 12.5 shows the extent of the sites affected by uranium mining and milling in the Czech Republic.

The state-owned enterprise DIAMO is now responsible for a large-scale closure and contaminated site clean-up programme in the Czech Republic, including technical, social and environmental aspects. The first decommissioning and remediation of exhausted mines were undertaken in the 1950s, but these works are often now considered inadequate. As part of the policy of continuing improvement of the environment, the government is funding the remediation of these sites. It is expected that these activities will have to continue until about 2040 and are estimated to cost approximately 3 billion euros.

The remediation of tailing ponds involves dewatering and treatment of contaminated waters from the tailing impoundments. The tailings are protected against infiltration of ambient precipitation, as well as against groundwater inflow. The final aim of the remediation is to blend the remediated sites into the landscape.

The research activities in the nuclear field in the 1950s also created sites, which have to be cleaned up. One of these sites is NRI in Rez. Over more than 55 years of activities in the nuclear field, a lot of contaminated

Table 12.5 Uranium mining waste dumps and milling tailings in the Czech Republic

Localities	Waste dumps	Tailings	
	Volume (thousand m^3)	Extent (ha)	Volume (thousand m^3)
Straz pod Ralskem	1,137	187.0	19,236.0
Rozinka	3,290.0	90.1	9,827.4
Mydlovary		292.7	23,969.0
Pribram	30,072.0	44.1	238.3
West Bohemian Region	2,125.0	20.1	2,798.0
Jachymov	14,382		
Others	7,416		
Total	58,422	634.0	56,068.7

Source: Tomas (2009).

materials, technologies and structures have been generated. Two areas are especially in need of remediation (Podlaha, 2007):

1. Decommissioning of old obsolete facilities (e.g., decay tanks, liquid RAW storage tanks, old RAW treatment technologies, special sewage systems).
2. Processing of RAW from operation and dismantling of NRI nuclear facilities.

The character of the environmental liabilities is very specific and requires special remediation procedures, development and/or purchase of a number of tools for fragmentation, decontamination and disposal of radioactive wastes. The first phase of remediation activities started in 2003 and will be finished in 2012. All activities are paid for by the Czech state. The first phase will cost approximately 40 million euros.

12.5 Problematic cases and lessons learned

Problems of RAW management in Eastern Europe vary from country to country. The management of RAW systems in both the Czech and Slovak Republics from operation and decommissioning of nuclear facilities was influenced by the Soviet design concept of waste management of WWER reactors, which allowed for the fact that virtually all RAW will be stored until decommissioning of the NPP. A great disadvantage of this design was also the fact that various sources of wastewater were mixed and therefore recycling of separate wastewater is complicated and difficult. The consequence of this design is that usually much higher amounts of waste have to be disposed of at the low- and intermediate-level waste disposal facilities such as at Dukovany or Mochovce, unlike in Western reactor designs. Both in the Czech and Slovak Republics, however, special programmes focusing on reduction of generated waste have been launched.

In the Czech and Slovak Republics the main conditioning technology for operational liquid waste from NPPs selected by the end of the 1980s was bituminisation. This technology is associated with many problems such as maintaining a suitable pH of wastewater concentrates or flammability of bitumen, necessitating conducting fire hazard tests prior to waste conditioning. Another problem in both countries is finding suitable sites for deep geological repositories. In the Czech Republic, the process started by selection of sites only according to geological criteria. It turned out, however, that socio-economic aspects are equally as important as geological criteria for selecting suitable sites. The biggest problem for finding suitable sites is the rejection of these sites by local communities and non-governmental organisations.

In the Czech Republic problems with remediation of sites after uranium mining and milling remain. It may be that the sites will be cleaned up so that they will not significantly endanger people and the environment, but only after spending large amounts of money and it is unlikely that most of these sites will be open for free, unrestricted use in future. They will have to remain under some institutional control, probably indefinitely. They will require constant monitoring and periodic assessment, and, if required, maintenance.

12.6 Future trends

As mentioned earlier, as the Czech Republic, Slovakia and Poland are planning the construction of new NPPs, this must go hand in hand with development of waste management systems compatible with available equipment and storage and disposal facilities. Experience gained at existing NPPs yielded a number of findings, which can be used to build new waste management systems for these new NPPs. Firstly, it was found that any improvement to waste management systems after start-up of operation of the NPP is very costly and sometimes impossible. This concerns primarily the management of liquid waste. Secondly, it was determined that the bituminisation technology can provide good waste form properties, but the technology itself requires additional measures to reduce the risk of fire and relatively complex waste liquid pre-treatment to prevent the crystallisation of boric acid.

Both the Czech and Slovak Republics and Poland have launched several scientific projects concerning the development of new waste management technologies (Vokál *et al*., 2007, Hanusik *et al*., 2008, Noferi, 2009). The results suggest that implementation of new waste management technologies (new liquid waste treatment systems, new conditioning technologies such as polymer encapsulation or embedding of waste in ceramic materials) could significantly improve the waste management systems, but their implementation will require much effort and money.

In the Czech Republic a programme of deep geological disposal of SF is under way and in the Slovak Republic and Poland is under preparation, but all face many problems connected primarily with finding acceptable sites for location of the repository.

12.7 Sources of further information

Further information can be found primarily in Czech, Slovak and Polish National Reports prepared under the Joint Convention on the safety of spent fuel management and on the safety of radioactive waste management (Czech National Report, 2008, National Report of Slovak Republic, 2008, National Report of Poland, 2008). A wide range of information is included

in a variety of IAEA reports. In particular, it is possible to find a lot of information on waste management systems in Eastern European countries in the reports dealing with WWER waste management system or remediation of contaminated sites after uranium mining and milling activities (IAEA, 1995, 2000, 2004, 2005, 2006).

It is also possible to find summaries of information on waste management systems in Eastern European countries in European reports compiled before accession of these countries to the European Union (EC DGXI, 1999, 2000).

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14

Germany: experience of radioactive waste (RAW) management and contaminated site clean-up

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Abstract: The Federal Republic of Germany has committed to the complete phase out of nuclear energy production by 2022. Considerable effort has been expended on developing deep geological repositories for radioactive waste (RAW) associated with energy production and industry. Three such repositories, Asse, Morsleben and Konrad for wastes with negligible heat generation exist in Germany. Asse and Morsleben are both being closed in accordance with the German Atomic Energy Act, while Konrad has been licensed to receive waste and is currently being constructed. An exploratory facility for the deep geologic disposal of heat generating radioactive wastes is located at Gorleben, Lower Saxony. Related repository design studies continue to progress and specialized full-scale waste handling and emplacement equipment has been designed and tested.

Key words: nuclear energy phase-out, German Atomic Energy Act, waste canisters, interim storage, deep geological repositories.

14.1 Introduction

Germany is the fifth largest economy in the world and the largest within the European Union (US CIA, 2011). Germany is also the largest generator of electrical energy in the European Union. In 2010 electrical energy generation was 622.5 TWh, of which 22.5%, or 140.6 TWh, was produced from nuclear power generation (AGEB, 2010), approximately 57.6% from fossil fuels sources, 15.9% from renewables, and 4% from other sources (AGEB, 2010, 2011; ENS, 2011). Prior to the Daiichi Power Plant nuclear incident in Fukushima, Japan, in March 2011, Germany operated 17 nuclear power plants (NPPs) with a total net capacity of 20.49 GW (ENS, 2011); six are boiling water reactors (BWRs) and eleven are pressurized water reactors (PWRs). In response to the incident in Fukushima and in the face of an increased anti-nuclear atmosphere in German society, the federal government immediately ordered the removal of 8 NPPs from service in response to the unfolding crisis and committed to a phase-out of nuclear energy by 2022. As a direct consequence, the actual contribution of nuclear energy to

Germany's electrical power generation is currently 11%. Prior to the Fukushima incident, Germany had planned on extending the life of the NPPs by an average of 12 years to 2036.

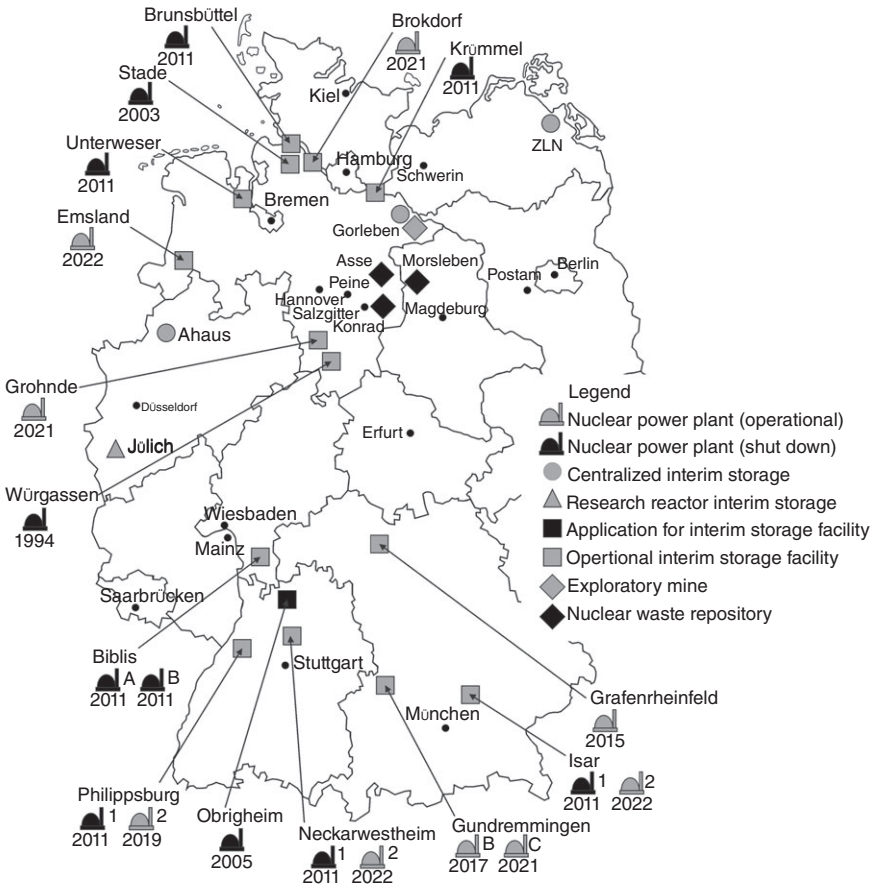
German nuclear energy production peaked in 2001 at 171.3 TWh with 19 nuclear power plants in operation at the time. Since then two power plants, Stade and Obrigheim, have been shut down and are currently being decommissioned. Seventeen further power and prototype reactors have either been shut down and are in the process of decommissioning or have been completely decommissioned (BfS, 2011a). The decommissioned plants include all of the Soviet designed VVER (water-water energetic reactor) constructed and operated by the former German Democratic Republic (GDR); the two prototype thorium high-temperature reactors; the prototype fast breeder SNR-300 nuclear reactor near Kalkar, Germany; as well as several BWRs and PWRs. The locations of Germany's key nuclear sites are shown in Fig. 14.1.

In addition to nuclear reactors used for power generation, a total of 37 research reactors have been constructed and operated in Germany. The majority of the former research reactors operated at very low to low power generation levels (i.e., in the range of 1.0×10^{-7} to 1.0 MW). Of the remaining eight research reactors, five also operate at these very low levels. Germany also maintains three nuclear fuel cycle facilities, while 11 further facilities are either in decommissioning or have been completely decommissioned (BfS, 2011a, 2011b). Since the end of the Second World War Germany has refrained from developing nuclear military capabilities.

Since 2002 the German federal government has been officially committed to a phase-out of nuclear electric power generation. Although the operational lifespan of Germany's NPPs was initially extended by the current government, the events at the NPPs in Fukushima, Japan, have resulted in a change of course by the federal government and the phase out has been expedited. Germany will now complete its phase out from nuclear energy production by 2022.

14.1.1 Overview of German Atomic Energy Act

In Germany, the regulatory framework for nuclear facilities and related radioactive waste (RAW) management is based on a hierarchy of acts, ordinances, safety rules and guidelines, and is consistent with pertinent European Law. The fundamental law governing all German nuclear facilities is the 'Law Over the Peaceful Use of the Nuclear Energy and the Protection against their Dangers' of 1959 as amended, also known as the Atomic Energy Act (Atomgesetz – AtG). In its original form, the AtG provided the basis for licensing and regulating nuclear facilities and the



14.1 Map of major German nuclear installations (ZLN – Zwischen Lager Nord (Interim Storage North); dates indicate year of NPP shut down or planned shut down). Source: Provided by the German Company for the Construction and Operation of Waste Repositories (DBE), Peine, Germany.

effects of ionizing radiation as well as for the handling and disposal of nuclear wastes. However, under the coalition government elected in 1998 between the Social Democratic Party (SPD) and the Green Party, the AtG was amended, based on agreements negotiated between the federal government and the major electrical utilities, to promote the phase-out of electricity production from nuclear energy (AtG §1). The amendment went into effect in April 2002. A key component of the AtG, as amended, was the implementation of a lifetime cap of 2,623.31 TWh on all operating nuclear power plants, which translated to an average 32-year total operational life for the existing facilities. In December 2010 the AtG was again amended

by the then-current coalition government to increase the cap by 1,804.278 TWh, thus extending the lifetime of the 17 remaining nuclear power plants by an average of 12 years (AtG §7(1a); Annex 3), and to include the provision for land expropriation contingencies for the study and development of nuclear waste repositories, if needed (AtG §9d and 9e). The AtG further assigns regulatory responsibilities between the federal government and the German Länder (Federal States) and makes provisions for the delegation of activities to third-party entities (AtG §9a(3)).

The recent incidents involving the Daiichi Power Plant in Fukushima, Japan, following the earthquake and tsunami of 11 March 2011, resulted in a freeze on plans by the German federal government to extend nuclear power plant operating life as announced by Chancellor Merkel on 14 March 2011. On 30 May 2011, the German federal government announced plans to leave off-line the seven oldest NPPs, which were immediately powered down after the disaster. An additional reactor, which had previously been shut down for maintenance purposes, will also remain off-line. The remaining operational NPPs will be shut down by 2022. The corresponding changes to the AtG were ratified by the German Parliament (Bundestag) and the Federal Council (Bundesrat) and incorporated into the AtG by German Federal Law Gazette 2011 Part I No. 43. The changes to the AtG became effective as of 6 August 2011.

Independent of the phase-out of nuclear energy in Germany, ensuring human and environmental protection requires a permanent solution for the RAW that has been and will continue to be generated. The disposal of these wastes in geological repositories is the only solution that ensures the protection of both humans and the environment for future generations.

14.1.2 Responsibilities under the German Atomic Energy Act for waste disposal

According to the AtG (§9a) the federal government is responsible for the final disposal of RAW in Germany. Within the federal government, the Federal Ministry for the Environment, Nature Conservation and Nuclear Safety (Bundesministerium für Umwelt, Naturschutz und Reaktorsicherheit, BMU) together with its devolved office the Federal Office for Radiation Protection (Bundesamt für Strahlenschutz, BfS) are responsible for the siting, planning, facility-related research and development, exploration, construction, operation, and subsequent decommissioning and closure of repositories for RAW. The Federal Ministry of Economics and Technology (Bundesministeriums für Wirtschaft und Technologie, BMWi) is responsible for developing federal energy policy and application of pertinent aspects of Federal Mining Law. The Federal Institute for Geosciences and

Natural Resources (Bundesanstalt für Geowissenschaften und Rohstoffe, BGR), a subordinate authority of the BMWi, deals with the primary geoscientific questions surrounding the final disposal of RAW. Licensing and license authorization falls under the province of the pertinent Federal State ministries; however, to ensure the uniform implementation of the AtG, these ministries are subject to supervision under the BMU. The BfS is the licensee for geological disposal facilities and is empowered by the AtG to establish final repositories for radioactive wastes (AtG §23(1)2).

The BMU is the authority for all nuclear safety and radiation protection issues. In this field it has the competence to issue directives and to supervise the legality and expediency of the acts of authorities responsible for enforcing the Atomic Energy Act and the Radiation Protection Ordinance. The BMU develops the regulatory framework; it sets the regulatory dose levels on allowable radiation exposure for disposal facilities, and defines the performance periods over which these are to be assessed for disposal facilities. The BfS, under authority of the BMU, implements federal administrative tasks in the field of radiation protection and in the management of radioactive waste, in particular with respect to the construction and operation of disposal facilities. In accordance with §9a, section 3, of the AtG, the Federal Republic of Germany, represented by the BfS, commissioned the German Company for the Construction and Operation of Waste Repositories, (Deutsche Gesellschaft zum Bau und Betrieb von Endlagern für Abfallstoffe mbH, DBE), as a third party to design, construct and operate federal repositories for RAW.

For waste disposal purposes, the BMU defines two categories of nuclear waste: waste with negligible heat generation or heat-generating waste. Since the 1960s German federal policy has been that all solid or solidified forms of RAW subject to control by the AtG are to be disposed of in deep geological formations. Although all of these wastes will be disposed of in deep geological formations, a differentiation is made as to which types of wastes can be disposed of in a specific facility.

The BMU promulgated new safety requirements for heat-generating waste in September 2010 (BMU, 2010). The safety requirements provide an annual dose limit over a one million year performance period of 0.01 mSv for probable and 0.1 mSv for low probability future developments (BMU, 2010, Section 6.2 and 6.3). These limits are significantly lower than the IAEA recommended dose constraint of not more than 0.3 mSv for geological disposal facilities after closure (Safety Series No. 115). Performance criteria for waste with negligible heat are governed by §47 of the Ordinance on the Protection against Damage and Injuries Caused by Ionizing Radiation (Verordnung über den Schutz vor Schäden durch ionisierenden Strahlen, StrlSchV) and developed separately in site-specific plan approval documents in accordance with AtG §9a and 9b.

14.2 Sources, types and classification of nuclear waste

Internationally, several different systems have been developed for the classification of radioactive wastes. These are generally based on half-life, activity levels, origin or source, or the degree of isolation required. In general, low level waste (LLW) contains radionuclides with low activities and short half-lives and generates no heat; some systems differentiate a subcategory for very low level waste (VLLW). Intermediate level waste (ILW) may contain radionuclides with low to intermediate activities and short to long half-lives, generating no to negligible heat. High-level waste (HLW) contains radionuclides with high activities, long or short half-lives or both, and generates heat (Rempe, 2007).

In Germany, as mentioned earlier, the BMU defines nuclear waste for disposal purposes based on its heat generating capacity, as either waste with negligible heat generation or heat-generating waste. In the German system waste with negligible heat generation consists of VLLW, LLW and ILW, while waste classified as heat generating consists of both spent nuclear fuel (SNF) and HLW. In accordance with federal policy as promulgated by the BfS, both waste types are to be disposed of in waste-specific deep geological repositories (BfS, 2011c). Construction, operation and closure of a repository must be approved according to the Atomic Energy Act (AtG §9b) as part of a planning approval process.

The major sources of radioactive wastes in Germany are associated with nuclear fuel cycle activities, power generation, research facilities, the reimportation of HLW associated with the reprocessing of SNF in the United Kingdom and France, decommissioning of the various nuclear facilities, and the use of radioisotopes in medical, research and industrial applications. Other materials, primarily associated with the decommissioning of nuclear facilities, which are either not radioactive or only weakly radioactive, can be released from nuclear regulatory control by permit providing applicable regulatory conditions are met (Chapter 2, Section 9, §29 of StrlSchV). The BfS estimates that a total of approximately 290,000 m^3 of waste with negligible heat-generating capacity will require disposal (BfS, 2011d). Of these, approximately 161,000 m^3 of the waste is expected from decommissioned NPPs by 2080 (BfS, 2011c). The current inventory of heat-generating nuclear waste requiring geological disposal in Germany as of 31 December 2010 is given in Table 14.1.

14.3 Radioactive waste (RAW) management strategies: history and developments

Germany is a contracting party to the Joint Convention on the Safety of Spent Fuel Management and on the Safety of Radioactive

Table 14.1 Inventory of heat-generating radioactive waste

Waste form	Quantity (in Mg heavy metal or m^3)
Estimated total SNF after phase-out is completed	11,100 Mg
SNF in reprocessing by AREVA at La Hague	5,379 Mg
In reprocessing by BNGS at Sellafield	851 Mg
Various German and foreign locations[a]	440 Mg
Unconditioned waste (recyclable and raw wastes)	3 m^3
Intermediate level wastes[b]	1,251 m^3
Conditioned waste	674 m^3

[a] Includes SNF, as of December 2010 located at: Reprocessing Plant Karlsruhe (WAK); Eurochemie in Mol, Belgium; Central Holding Storage for SNF (CLAB) in Sweden; Reprocessing and storage facilities in the former USSR; and at NPP Paks in Hungary.
[b] Includes unused fuel from the thorium high temperature reactor (THTR).
Source: BfS (2011c).

Waste Management. Waste management legislation in Germany is based on European law, German federal law, and regional state laws. In accordance with the AtG, waste producers are committed to avoid or reduce the generation of radioactive waste to the greatest extent possible. Ownership of the waste is retained by the producer until such time as it has been accepted for final disposal in an approved geological repository (AtG §9a).

As previously discussed, all radioactive waste in Germany, subject to the controls of the AtG will, in accordance with federal policy, be disposed of in a suitable deep geological repository. The specific requirements that a repository must meet are determined based on the heat-generating capacity of the waste destined for disposal and the isolation requirements associated with the waste. These requirements are specified in the site licensing documents as required by the pertinent German laws. Pending disposal, waste related to power generation is managed in secure on-site or near-site interim storage facilities at the expense of the waste producer. For all other wastes, particularly those originating from radioisotope applications in industry, universities and medicine, the Federal States are responsible for constructing and operating regional interim storage facilities (AtG §9a(3)).

Germany was one of the first nations to initiate serious efforts in developing strategies and techniques for deep geological disposal of RAW. The German government policy on deep geological disposal for all radioactive wastes can be traced back to 1960 when the former German Atomic Commission unilaterally rejected the idea of surface disposal for these wastes. In 1967, Germany initiated a pilot test-bed geological disposal facility for

low and intermediate level wastes (LLW and ILW) at the former Asse salt mine. The facility was the first attempt at developing a prototype repository for the storage of nuclear wastes by any nation (Fisher, 1978). In 1971, the former GDR (East Germany) began disposing of LLW and ILW wastes in the rock salt mine Bartensleben near Morsleben, Saxony-Anhalt. Waste storage practices at Asse ceased in 1978, while waste disposal practices at Morsleben continued uninterrupted until 1991 and again from 1994 until 1998. Currently the only facility licensed in accordance with the AtG for the disposal of negligible heat-generating wastes in Germany is being constructed in the former iron-ore mine at Konrad.

With respect to the geological disposal of heat-generating wastes, i.e., HLW and SNF, Germany was one of the first nations to initiate serious efforts in developing a permanent deep geological repository for heat-generating wastes, and by 1977 had selected the salt dome at Gorleben for investigation regarding the suitability of the formation for hosting a potential repository for HLW and SNF.

14.3.1 Reprocessing SNF and related HLW

Germany currently operates a once-through fuel cycle. Although initial intentions were for a closed fuel cycle, strong public opposition and economic concerns led to the abandonment of plans for a reprocessing facility in the Bavarian town of Wackersdorf in 1988. Until 1994 utilities were obliged to reprocess SNF in order to recover usable materials for recycling into new fuel assemblies. However, because Germany never fully developed the capability, most of the reprocessing of SNF was contracted to facilities in France and the United Kingdom. Only a small amount of fuel was reprocessed in Germany at the Karlsruhe reprocessing plant (Wiederaufarbeitungsanlage Karlsruhe, WAK). Between the commissioning of the WAK in 1971 and its shut-down in 1990, about 200 tonnes of irradiated fuel were reprocessed at the facility (EWN Gruppe, 2010). Federal policies began to change between 1994 and 1998 when both reprocessing and direct disposal were equally acceptable to the government. As part of the agreement negotiated between the SPD–Green coalition federal government and the nuclear utilities, it was agreed in 2001 that SNF would be disposed of directly and foreign shipments for reprocessing SNF would no longer be allowed after mid-2005 (subsequently codified into the AtG §9a).

As of 31 December 2010, 97 casks of type CASTOR® HAW 20/28 CG or similar with vitrified HLW were being stored at the interim storage facility for heat-generating waste at Gorleben. An additional 33 casks of vitrified HLW will be shipped from France and the United Kingdom associated with reprocessing of German SNF (BfS, 2011c). Eleven of the casks were returned from the French reprocessing facility in La Hague by the end of 2011 and

21 will be returned from the reprocessing facility at Sellafield in the United Kingdom by the end of 2017. By contractual accord, LLW and ILW generated as a by-product of reprocessing will remain at the foreign facilities. As an offset, approximately 5% additional canisters with vitrified HLW are included in the waste being returned to Germany for final disposal. Additionally, by the end of 2024, the final shipment of approximately 150 CASTOR®-type casks containing high-pressure compacted waste will be returned to Germany from La Hague.

14.3.2 Waste transportation and waste containers

Every year hundreds of thousands of packages containing radioactive materials, primarily of medical origin, are transported in Germany. However, only a small number of nuclear transports are conducted for materials associated with either the nuclear fuel cycle or the decommissioning of nuclear facilities.

The transport of heat-generating waste in Germany, specifically with respect to HLW being returned from France and the United Kingdom, has faced very strong public and political opposition. Waste transports are regularly disrupted by protesters and a significant police presence is generally required. Partially as a result of the protests regarding transportation of waste in Germany, storage of SNF at centralized locations has been replaced by decentralized interim storage facilities located at the various NPP sites.

Germany has developed two primary systems for the transport and/or storage of heat generating waste: the CASTOR® and POLLUX® systems (GNS, 2011a). The CASTOR® family of containers is licensed for transport and interim storage, while the POLLUX® family of containers is intended for final disposal. Both containers were developed by the Gesellschaft für Nuklear-Service mbH (GNS). Additionally, the GNS has developed the BSK 3 container concept for borehole disposal of SNF in salt, as well as specialized containers for waste products from research reactors.

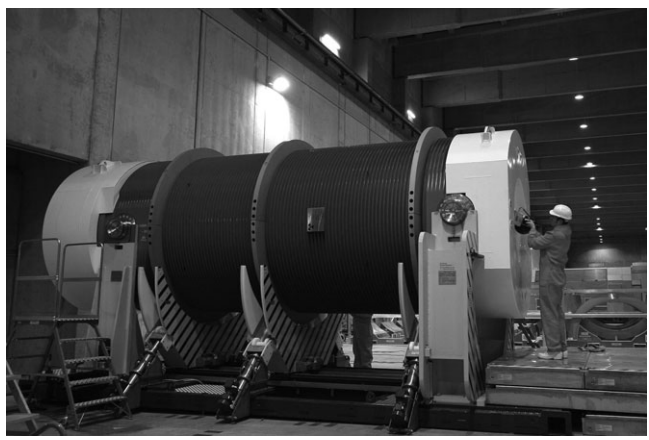
CASTOR® nuclear waste container systems

For transportation and interim storage, Germany developed and licensed the CASTOR® cask system. This system includes a number of different variants based on the intended contents. However, in general all CASTOR® containers consist of a double-shell design sealed with two separate end-cap sealing systems. The cask body consists of a large cylindrical 30–40 cm thick-walled casing made of ductile cast iron steel. The interior of the CASTOR® container is nickel plated. For neutron moderation, axial boreholes are distributed uniformly in the cask wall to accommodate moderator rods. The

bottoms of the containers are sufficiently thick to provide gamma and neutron shielding. The lid system consists of a double-barrier sealing system upon which a third protective cover is placed during storage. During transportation, both the lid and the bottom ends are protected by large steel-plate shock absorbers. The exterior design of most of the CASTOR® containers incorporates cooling fins designed to radiate access thermal energy from SNF or HLW that are still generating heat. The casks are loaded under water. Today the most widely used CASTOR® containers are the types CASTOR® V/19 (for the contents from 19 spent fuel assemblies used in pressurized water reactors) and CASTOR® V/52 (for the contents from 52 spent fuel assemblies used in boiling water reactors). These containers are approximately 6 m long with a diameter of approximately 2.5 m and weigh approximately 125 tonnes when fully loaded (GNS, 2011b; BAM, 2010). Feasibility studies are currently under way regarding the potential use of the CASTOR® cask for geologic disposal. A typical CASTOR® is shown in Fig. 14.2.

POLLUX® nuclear waste container systems

The POLLUX® was specifically developed for the geological disposal of SNF, but is also suitable for interim storage and transportation if required. A single POLLUX® can hold ten irradiated fuel rods from a PWR or 30 fuel rods from a BWR. It has a diameter of 1.6 m, a length of approximately 5.5 m, and a weight of 65 tonnes when loaded. The container is a double-shell design with an internal container to accommodate the fuel rods from



14.2 CASTOR® HAW28M container at the centralized interim storage facility in Gorleben. Source: Provided by GNS Gesellschaft für Nuklear-Service mbH, Essen, Germany.

SNF assemblies, which are separated by a neutron moderator, and an external shield container made of spheroidal graphite (SG) iron (GNS, 2011b; Diersch *et al.*, 1993). Figure 14.3 shows a POLLUX® container being hoisted into place for a 9 m edge-on drop test.

BSK 3 container

The BSK 3 container concept is intended for the direct borehole disposal of SNF in salt rock. The container can accommodate the fuel rods from three PWR or nine BWR fuel assemblies. The container is unshielded and has a maximum diameter of approximately 440 mm. The diameter of the BSK 3 container was selected so that the SNF could be disposed of in the same type of borehole that is planned for the disposal of HLW casks. A variation BSK 3 concept referred to as the 'Triple-Pack' was developed by DBE Technology GmbH to contain three HLW casks with a diameter of 430 mm. Using the Triple-Pack and BSK 3 concepts for borehole disposal would optimize the use of handling and emplacement equipment requirements.

Containment of wastes with negligible heat-generating capacity

Liquid LLW and ILW wastes are either evaporated or mixed in cement, while solid wastes are crushed, incinerated, compacted or cemented

14.3 A POLLUX® container being lifted into position for a 9 m drop test. Source: Provided by the German Federal Institute for Materials Research and Testing (BAM), Berlin, Germany.

beforehand. All waste types are packaged in standardized and approved containers after processing. Cylindrical concrete containers are generally used for solidified waste, while unconditioned waste is sealed in iron waste containers with welded lids. LLW and ILW currently in interim storage will be disposed of in the Konrad facility. Existing waste at Morsleben will be considered as geologically disposed upon completion of the facility closure. No final decision has been made as to the ultimate disposition of waste currently stored at the Asse facility.

14.3.3 Contaminated site clean-up experience

Contaminated site clean-up in Germany is primarily associated with the decommissioning and dismantling of former nuclear facilities. Germany has considerable experience in the decommissioning and dismantling of nuclear facilities. The preferred decommissioning strategy in Germany is the immediate dismantling of facilities as opposed to safe enclosure. The BMWi chose this option for the Greifswald nuclear power station, where five reactors had been operating. The former 100 MWe Niederaichbach NPP site was declared safe for unrestricted agricultural use in mid-1995. In addition to the Niederaichbach NPP, the Karlstein superheated reactor has also been fully decommissioned and returned to a 'green-field' state. Seventeen additional NPPs are at various stages of decommissioning and dismantling. Additionally, 28 research reactors and 11 facilities associated with the nuclear fuel cycle have either completed decommissioning or are currently being decommissioned. As mentioned previously, as a result of the incident at the Fukushima nuclear station, the plans for the complete phase-out of German nuclear power production are being accelerated and considerable effort will be required with respect to the dismantling and decommissioning of the remaining German NPPs.

Decommissioning of nuclear facilities in Germany is based on the polluter-pays principle. With the exception of NPPs associated with the former GDR, the electric utilities are responsible for all current and former operational NPPs. Responsibility for NPPs associated with the former GDR was transferred to the Federal Ministry of Economics and Technology (Bundesministeriums für Wirtschaft und Technologie, BMWi) in accordance with the German Reunification Treaty. The Federal Ministry of Education and Research (Bundesministerium für Bildung und Forschung, BMBF) is responsible for the management and decommissioning of nuclear research facilities.

Germany's first commercial nuclear reactor, the 250 MWe Gundremmingen-A unit, operated from 1966 to 1977; decommissioning started in 1983. In 1990, using specifically developed underwater cutting technologies, dismantling of the highly contaminated portions of the facility

began. Gundremmingen-A demonstrated that decommissioning could be undertaken safely and economically without long delays. Most of the metal from the facility was also successfully recycled (Hore-Lacy, 2009).

Decommissioning of the 17 currently operating and recently shut down reactors is expected to produce some 115,000 m^3 of decommissioning wastes (WNA, 2011). Decommissioning wastes which fall under the control of the AtG with negligible heat-generating capacity will be disposed of at the Konrad repository once the facility becomes operational. Heat-generating waste will remain at interim storage sites pending the availability of a final repository.

Prior to German reunification in 1990, the former GDR in conjunction with the former Soviet Union developed the world's third-largest uranium mining province operated by the joint German–Soviet company Wismut SAG. Operations continued from 1946 to 1990 for a total production of 220,000 tonnes of uranium. A significantly smaller uranium ore mining operation was also conducted in western Germany near Ellweiler. Germany no longer mines uranium currently and all uranium used in fuel production is imported. The sites have largely been restored to green-field status (Wismut GmbH, 2011; MUFV, 2011).

14.3.4 Interim storage

Heat-generating wastes are placed in interim storage for decaying and cooling at a total of 15 separate facilities in Germany pending their final disposal in a future geological repository. Germany maintains three centralized storage facilities at Gorleben, Ahaus, and the Interim Storage North (Zwischenlager Nord, ZLN) facility on the site of the former NPP Greifswald. Twelve additional decentralized facilities are located adjacent to the various nuclear power stations. These facilities are subject to the regulatory authority of both the federal government as well as the local state governments.

The 2002 amendment to the Atomic Energy Act committed NPP operators to establish interim decentralized storage facilities for SNF resulting from plant operations. As a result, current wastes being generated are stored at the 12 decentralized locations. The facilities became operational and began accepting waste in 2006 and 2007. A thirteenth facility is currently in the licensing process. Spent fuel from earlier NPP operations is stored at the centralized interim storage facilities. HLW returned from reprocessing in France and the United Kingdom is stored at Gorleben. The decentralized interim storage facilities generally consist of surface structures made of reinforced concrete (at the Neckarwestheim site storage tunnels are used). SNF is stored at these locations in CASTOR®-type transportation and storage casks.

Wastes classified as RAW with negligible heat generation are produced in association with research institutions, the nuclear energy industry, decommissioned nuclear facilities, the former reprocessing plant at Karlsruhe, and various other state and industrial activities. These wastes are in the form of LLW and ILW and are stored at numerous interim storage facilities in Germany. The facilities are subject to the regulatory control of the Federal States where they are located. Upon completion of the Konrad repository for wastes with negligible heat generation, these wastes will be transported and permanently disposed of at the licensed facility.

14.3.5 Waste disposal strategies

While many countries have opted for near-surface or surface disposal for LLW and ILW, Germany has pursued deep geological disposal for all RAW subject to the controls of the AtG. Additionally, it has been German federal policy since the early 1960s that deep geological disposal offers the best possible isolation of the wastes. Deep geological disposal is seen as particularly beneficial over surface or near-surface disposal with respect to the avoidance of inadvertent human intrusion. To this end, even wastes with no or only negligible heat-generating capacity are, and will continue to be, disposed of in deep geological repositories.

Approximately 98.5% by volume of the nuclear waste generated in Germany is classified as waste with negligible heat-generating capacity. Because Germany has selected deep geological disposal for these wastes, it is necessary that sufficient underground volumes are located and that the costs associated with deep disposal are manageable without adversely affecting safety. The conversion of existing underground mines, assuming the facility can be determined to provide adequate isolation and safety, offers an alternative to the development of new purpose-built facilities for disposal of these wastes. As a result, Germany pursues two separate strategies for radioactive wastes disposal. For wastes with negligible heat-generating capacity (i.e., short-lived LLW and ILW), former mines, which have been extensively studied and retrofitted, are used for disposal purposes. However, before an existing mine can be used for geological disposal purposes, the facility must be thoroughly evaluated and appropriately designed to provide engineering alternatives where the original purpose of a mining facility may diverge from the safety and isolation requirements of a repository. For heat-generating waste, only a purpose-built facility in previously undisturbed geological formations is seen as appropriate.

Currently, Germany operates an underground exploratory facility in the Gorleben salt dome. The exploratory facility is specifically tasked with studying the suitability of the Gorleben salt dome as a potential repository host formation for heat-generating wastes. However, the site has not yet

been selected as a potential repository and a final decision on the site's suitability will depend on the results of additional investigations and positive findings from future safety assessments.

Germany currently has three existing facilities which are classified as geological repositories for the disposal of nuclear wastes with negligible heat-generating capacity: Asse, Morsleben and Konrad. Asse and Morsleben will both undergo closure as repositories in accordance with the AtG, but only Konrad was licensed for disposal under the AtG.

14.4 German nuclear waste repository projects

14.4.1 Asse II

At the same time that Germany was constructing its first NPPs, the government recognized that methods and technologies would need to be developed for the final geological disposal of related heat-generating wastes. The permanent disposal of these wastes in salt domes was seen as providing a promising option for the development of a HLW repository. To further investigate the ability of a salt-rock formation to serve as a potential repository host rock, the German Federal Ministry of Education and Research (Bundesministerium für Bildung und Forschung, BMBF) acquired the former Asse potash and rock salt mine in 1965 as a prototype facility for LLW and ILW disposal with strong emphasis on research and disposal technologies (BfS, 2011e). The facility was managed at the time by the GSF (Gesellschaft für Strahlen- und Umweltforschung mbH), a major research centre in Germany, which later became the Helmholtz Zentrum München (HZM). Management and operations of the Asse facility were conducted by the Helmholtz Zentrum München (HZM) until the facility was transferred to the BfS in 2009. From its initiation until 2009, Asse was regulated under German mining laws.

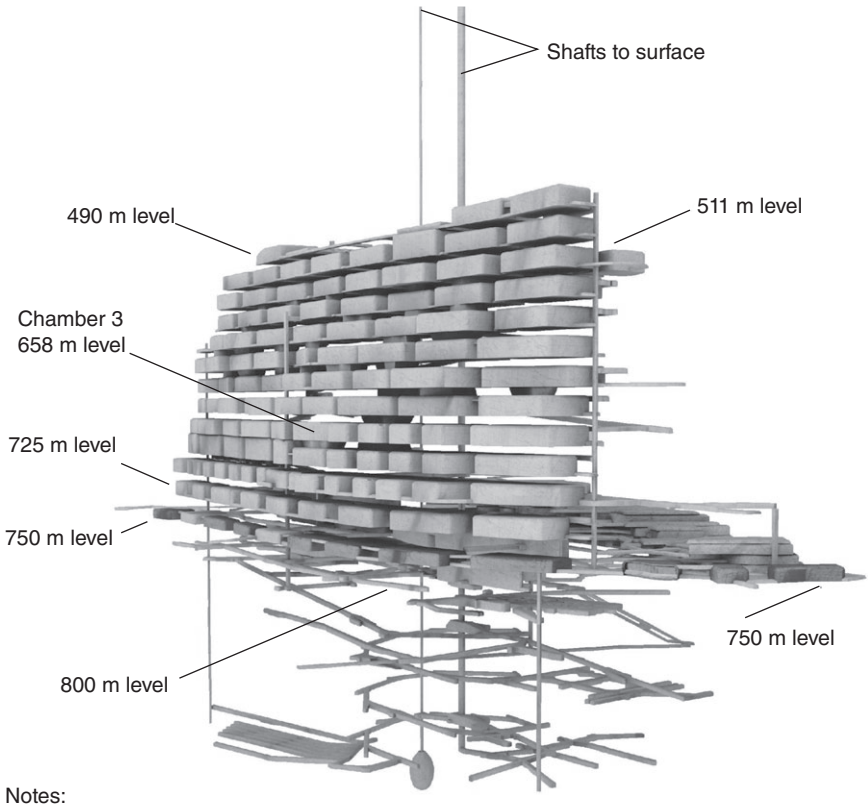
Research and experimental work on remotely handled ILW disposal started in the summer of 1972 and continued until waste disposal practices ended in 1978. From 1971 until 1978 the facility was also used to store a major part of the LLW and ILW produced in the Federal Republic of Germany. Altogether 125,787 drums and waste packages containing RAW were emplaced in the mine (BfS, 2011f). The layout of the Asse facility including chambers containing RAW is shown in Fig. 14.4.

The AtG was amended in 1976. The amendment implemented a licensing (i.e., plan approval) process for RAW storage and as a result waste storage practices were discontinued in 1978. At the time, as no additional RAW was being transferred to the site, German mining law continued to provide the legal basis for the operation of the facility as an underground research laboratory (URL). After phasing out of the disposal practices in 1978, the facility continued to be used as an underground research laboratory with

a major focus on the development of disposal technologies for heat-generating waste.

In 1995, after research and development came to an official end, backfilling of the former mining chambers in the southern flank of facility was initiated along with efforts to evaluate the long-term safety of the former mine. However, research was allowed to continue as long as related activities did not interfere with mine closure operations (Kappei, 2006).

Although the facility was initiated as a URL, because of the disposal practices that were conducted concurrent with research, the facility became



Notes:

(1) Intermediate level radioactive waste is stored in a chamber located on the 511 m level
(2) Approximately 10 m^3 of brine is collected daily from Chamber 3 on the 658 m level and pumped to a storage tank located on the 490 m level
(3) Low level radioactive waste is stored in multiple chambers located on the 750 m level
(4) Small quantities of brine are also collected from locations on the 725 and 750 m levels and pumped to storage tanks on the 800 m level
(5) The lower mine chambers were partially filled during previous operations prior to assumption of responsibilities by BfS

14.4 Three-dimensional representation of the Asse repository – view of the southern flank. Source: Provided by the German Federal Office for Radiation Protection (BfS), Salzgitter, Germany.

by default a repository. It has since been recognized that the operation and regulation of Asse under mining law did not provide an adequate regulatory framework to manage and close the facility. On 4 September 2008, the BMBF, the BMU and the Lower Saxony Ministry for the Environment and Climate Protection (Niedersächsisches Ministerium für Umwelt und Klimaschutz, NMU) jointly agreed that the facility would be closed under the Atomic Energy Act. On 1 January 2009, transfer of the facility to the BMU under management of the BfS was completed.

Despite legal and political issues surrounding the Asse facility today, considerable experience and information was gained during the period of its operation. This experience and the tests that were conducted at Asse resulted in improved waste handling practices and technologies, as well as an improved understanding of salt as a host rock and engineered barrier system behaviour. Several national and international research studies were conducted at the URL. Examples include the following:

- a cooperative research programme with the US Department of Energy examined brine moisture migration, thermal mechanical response of salt, and material corrosion studies;
- drilling optimization studies were conducted as part of the Commission of European Communities COSA Project;
- the longest running drift-scale thermal simulation study was initiated by the Karlsruhe Institute of Technology (KIT) and later expanded and finalized under the European Union sponsored multi-national BAMBUS I and BAMBUS II projects, which included dismantling and retrieval exercises.

The BAMBUS projects were the last significant research conducted at the facility (Bechtold *et al.*, 2004). Upon assuming operational responsibility for the Asse repository in January 2009, the BfS conducted a comparative study to assess the effectiveness of the various closure options. The options investigated included:

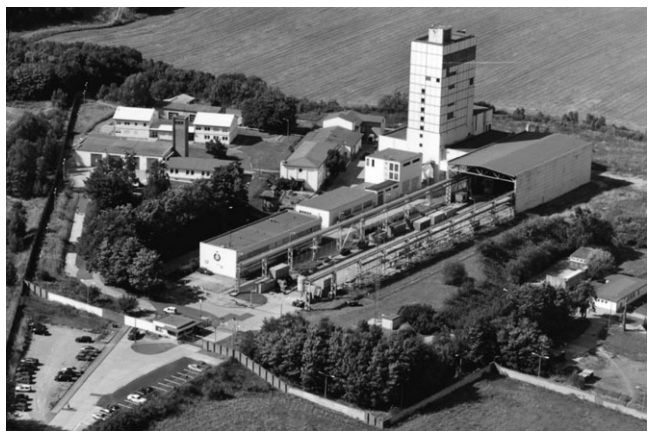
- retrieval: removal of waste from the mine for emplacement in another disposal facility
- relocation: construct and license a repository in deeper sections of the salt dome.
- complete backfilling: complete backfilling of all of the subsurface cavities with concrete and installation of sealing systems in shafts and drifts at appropriate geological intersections.

After evaluation of the result of the comparative assessment, published in January 2010 (BFS, 2010), the BfS selected retrieval as the preferred option for final closure of the facility and is currently in the process of elaborating technical processes and requirements to achieve this goal.

14.4.2 Morsleben

LLW and ILW originating from the operation of nuclear power plants, as well as from basic research, nuclear medicine and industrial applications in the former GDR was disposed of in the repurposed salt mine Bartensleben in Morsleben: the Morsleben Repository for Radioactive Wastes (Endlager für radioaktive Abfälle Morsleben, ERAM) from 1971 until German reunification (Fig. 14.5). The BfS became the licence holder upon reunification and DBE took over the operation of the facility as well as the task of designing any improvements and modifications through repository closure. After reunification, except for the period from 1991 to 1994 when emplacement operations were temporarily halted, disposal of low-level and medium-level radioactive waste with short-lived radionuclides continued until the Higher Administrative Court of Magdeburg issued an injunction on 25 September 1998 halting further disposal. On 12 April 2001, BfS committed to the permanent closure of the facility with no additional waste emplacement. During its period of operation from 1971 through 1998, a total of about 37,000 m^3 of RAW, including about 6,621 spent sealed radiation sources, was disposed of in the facility (BfS, 2011g).

The licence application for permanent closure, including the closure plan and the associated environmental impact statement, was initially submitted to the licensing authority, the Ministry of Agriculture and Environment of Saxony-Anhalt (Ministerium für Landwirtschaft und Umwelt Sachsen-Anhalt, MLU) on 13 September 2005. Revised documentation was resubmitted for review to the MLU in January 2009. The MLU completed its



14.5 Surface facilities at the Morsleben repository. Source: Provided by the German Company for the Construction and Operation of Waste Repositories (DBE), Peine, Germany.

review in July 2009 and the documents were submitted for public comment from 21 October 2009 to 21 December 2009.

Pending completion of the closure licensing process, work is ongoing to stabilize non-repository portions of the former mine. Specifically, extensive former mining activities in the central portion of the salt body raised significant concerns regarding the long-term stability of the subsurface openings. To address these concerns, backfilling of 27 former mine chambers with saltcrete was initiated on 8 October 2003. These operations were completed in February 2011. A total of approximately 935,000 m^3 of void volume have been filled in this manner. Final closure of the repository portions will commence after issuance of the closure licence (BfS, 2011h).

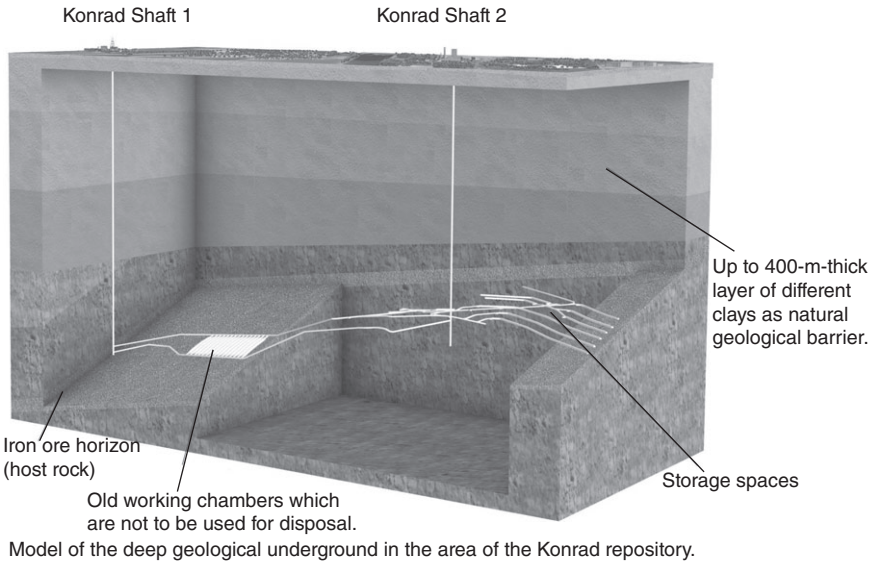
14.4.3 Konrad

In 1976 investigative efforts commenced at the former iron ore mine Konrad to assess its suitability as a repository for LLW and ILW. The original facility, consisting of two shafts, excavated in 1957 and 1960, respectively, was used to mine iron ore from an iron-rich very low permeable oolitic limestone formation between 1965 and 1976. A total of 6.7 million tonnes of ore were mined during this period.

The geological situation at Konrad offers favourable conditions for the disposal of RAW. The repository horizon is hydraulically isolated from overlying groundwater bearing formations. A more than 400 m thick and regionally widespread series of impervious clay, marl and mudstone layers cover the repository host rock and provide a geological barrier that, in conjunction with geotechnical barriers, will prevent radionuclides escaping into the biosphere (Fig. 14.6). Based on the favourable hydraulic conditions, considerations for repurposing the facility as a RAW repository were already initiated in 1975. In 1982 an application for the commencement of planning approval procedures was submitted.

After an almost 20-year licensing process, the Konrad facility was approved as a final repository in 2002. On 26 March 2007, the licence for Konrad was confirmed by the Federal Administrative Court. The ruling brought to a close all outstanding legal considerations and related judicial processes. Work on conversion of the mine began in May 2007 under the operational management of the BfS. The BfS is the licence holder and formal operator and manager of the facility, while DBE is assigned the responsibility for operating the facility and for the planning and construction of the repository.

The Konrad repository is the first final repository approved in Germany in accordance with the AtG. The facility is approved for the disposal of waste with negligible heat generation and has a licensed capacity for 303,000 m^3 of waste. Based on current waste forecasts, it is anticipated that a total of 290,000 m^3 of waste will be emplaced in the repository by 2050 (BfS, 2011d).



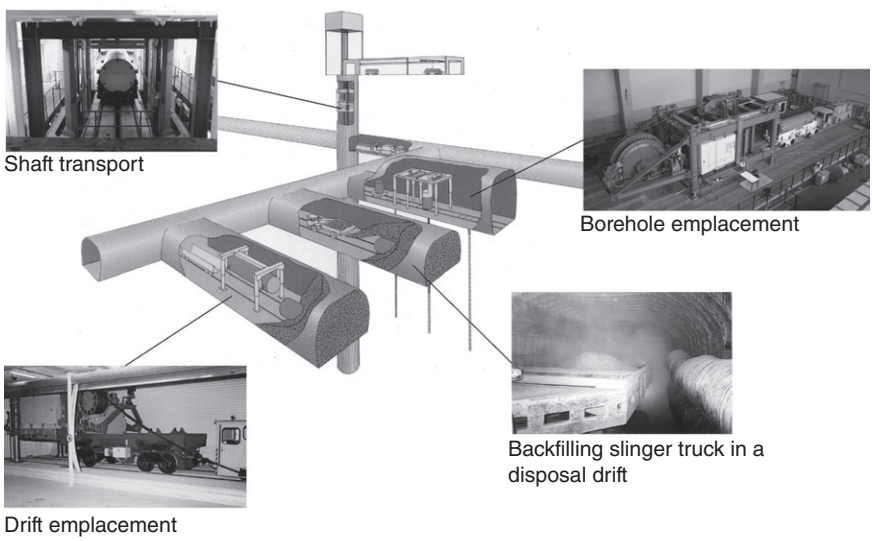
14.6 Three-dimensional representation of the Konrad Repository. Source: Provided by the German Federal Office for Radiation Protection (BfS), Salzgitter, Germany.

A total of 11 storage fields have been approved for the Konrad repository, although it is not anticipated that all of the available volume will be required. These fields will be constructed in the upper 800 m level of the repository. Refitting work is currently scheduled to be completed by 2019 after which waste acceptance operations will begin (BfS, 2011d).

14.4.4 Gorleben

The Gorleben facility is located in an undisturbed salt dome near the village of Gorleben approximately 100 km southeast of Hamburg, Germany. Following the selection of the Gorleben site in 1977 for investigation as a potential repository for heat-generating wastes, and the establishment of DBE in 1979, a comprehensive surface-based investigative programme was initiated to characterize the salt dome and the surrounding area of the site. Based on the positive indications from the surface investigations, an underground exploratory facility was designed and constructed by DBE in 1986 on behalf of the BfS. The Gorleben exploratory facility was intentionally designed to facilitate conversion to a repository, assuming subsequent investigations would continue to support the site's suitability. From 2000 to 2010, site characterization activities at Gorleben were suspended by the federal government as part of a moratorium agreement negotiated between the previous government and the nuclear industry. In October 2010 the moratorium expired and site characterization and licensing activities were restarted.

In the 1980s and 1990s, considerable effort was invested in investigating the Gorleben salt dome as a potential site for hosting a nuclear waste repository. The investigations supported the concept of rock salt as a host environment based on its very low inherent permeability and the self-healing nature of fractures due to the plastic response behaviour of the rock type. In addition to the subsurface research facility, many of the surface installations were also completed prior to the imposition of the ten-year moratorium. In the framework of research, development and demonstration activities, significant advances have been made with respect to prototype equipment development, including development of a shaft hoist system with a capacity to lift 85 tonnes, emplacement machines for both drift and borehole disposal, and equipment for backfilling disposal drifts. For these reasons, the facility at Gorleben is unique when compared to other international repositories in that much of the site characterization and surface infrastructure work was actually completed in the 1980s and 1990s. As a result, despite the moratorium, Gorleben remains one of the most technically advanced potential high-level RAW repository sites currently under consideration both in a national and international sense. Figure 14.7 shows the potential Gorleben repository concept and existing prototype equipment.



14.7 Gorleben repository concept with prototype shaft hoist, borehole emplacement machine, backfilling slinger truck and drift emplacement machine. Source: Provided by the German Company for the Construction and Operation of Waste Repositories (DBE), Peine, Germany.

Since the moratorium was lifted and research was recommenced, new safety requirements for the disposal of heat-generating waste, as well as requirements for retrievability have been published and are expected to be enacted. The performance criteria include the evaluation of repository safety for a one million-year period (referred to as the period of geological stability) at an annual effective exposure not to exceed 10 μSv for likely event scenarios and 100 μSv for less likely events (BMU, 2010). However, recent legal actions challenging key aspects of the operating licence for the Gorleben site investigation, submitted to the Upper Administrative Court of Luneburg, have resulted in the suspension of on-going subsurface research activities at Gorleben with immediate effect pending further judicial review.

A preliminary safety assessment (vorläufige Sicherheitsanalyse für den Standort Gorleben, VSG) that will provide a detailed evaluation of the potential suitability of the Gorleben salt dome as a repository host formation for the disposal of heat generating waste is currently being completed. The Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) is responsible for developing the VSG in collaboration with a team of contributing organizations, and is scheduled to be completed in 2013.

14.5 Problematic cases and lessons learned

For the disposal of waste with negligible heat generation, Germany has implemented a strategy of repurposing former mines as deep geological repositories. Although the deep geological disposal of these wastes is seen in Germany as strongly beneficial with respect to the protection of humans and the environment, in particular when compared to surface or near-surface disposal, implementation of the strategy has provided mixed results since the practice was initiated in the late 1960s and early 1970s. The two repositories associated with the early attempts at waste isolation, Asse and Morsleben, were not originally licensed subject to the AtG.

When Asse was originally commissioned as a research facility in 1965, before nuclear repositories were addressed in the AtG, it was subject to German mining law, while Morsleben, although commissioned as a repository, was subject to the nuclear and mining laws and regulations promulgated by the former GDR. The mining regulations under which Asse operated did not foresee the long-term safety requirements for closure of the facility. Unlike Asse, the legal framework under which the Morsleben facility was originally commissioned included rudimentary consideration for safety after closure of the facility. Upon German reunification, the former GDR laws applicable to Morsleben provided the basis for integrating the facility into the German repository programme. However, of the

three facilities, only the Konrad repository was designed and licensed subject to the full controls and regulation of the AtG.

The regulatory frameworks under which both Asse and Morsleben were originally commissioned are no longer applicable. Operational practices of the facilities as conducted at the time of their original commissioning, although consistent with the established law in effect at the time, would not be possible under the current German AtG. Therefore, although the strategy of repurposing former mines has had challenges associated with it, the success of the Konrad licensing process is a positive demonstration of the success of the strategy when implemented in accordance with an appropriate legal framework.

14.6 Future trends

The incident at the Fukushima nuclear power station in Japan has had a dramatic effect on the future of nuclear energy production in Germany. Prior to the incident, the German government had finalized plans for the extension of the operational life of the existing nuclear power plants. The extension of the operational life of the NPPs was seen by the current government as a key component of a broader energy plan meant to boost conservation, improve energy security, and move towards reliance on renewable sources of electricity, while remaining within the national goals for reducing carbon emissions.

Post-Fukushima, the federal government has reversed its previous policy. Seven older power plants and one which was undergoing repairs at the time of the Fukushima event have been powered down and removed from the electric grid. The government has expressed its intent that these power plants will remain off-line, while the remaining nine power plants will be phased out of operation in a staged approach to be completed by 2022. The impacts of these changes on energy policy and on the energy mix have not yet been fully evaluated.

Efforts to develop and close repositories will need to continue in order to address the nation's existing nuclear waste volumes and those of future generations. The reduced volume of expected waste will also result in a reduced repository capacity requirement and therefore presents an opportunity for re-examination of designs currently being considered, particularly with respect to designs under consideration for the potential repository at Gorleben. Although, the total volume of future waste generation is reduced by the recent policy changes, the need for waste storage capacity will likely increase more rapidly as NPPs are phased out earlier than previously planned.

The German federal government plans to draft legislation in the near future to align current laws governing the disposal of RAW with the new

Radioactive Waste and Spent Fuel Management Directive adopted by the European Union on 19 July 2011 (Enafact, 2011; EUROPA, 2011). Future priorities for the German repository programme include completion of the Konrad facility and closure of the Asse and Morsleben facilities, as well as completion of site characterization activities at Gorleben and finalization of a repository design and the associated safety case. Parallel to the finalization of activities for Gorleben, the federal government will also consider other potential host-rock options for the disposal of heat-generating wastes (BMU, 2011). On 11 November 2011, Minister of the Environment, Dr Norbert Röttgen, met with officials from each of the German federal states in a summit to discuss a path forward for selecting a repository site for heat-generating waste. At the summit an agreement was reached to draft legislation to provide a framework upon which to base a new site selection process. Consistent with BMU policy as identified in Safety Requirements Governing the Final Disposal of Heat-Generating Radioactive Waste (BMU, 2010), the search would be expanded to include all potential host-rock types, including salt, clay and granite. The original process, which focused solely on the Gorleben salt dome, has been sharply criticized for a number of years. However, Dr. Röttgen also stated that exploratory work on the Gorleben salt dome will continue and that the site has not been eliminated. The preliminary safety assessment for the Gorleben site (VSG) will be completed on schedule by the end of March 2013. Currently, negotiations between the federal government and the leaders of the opposition parties, specifically the Social Democratic Party and the Greens, are continuing. Depending on the outcome of these negotiations, significant changes could result to the German repository programme for the disposal of heat-generating waste.

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15

France: experience of radioactive waste (RAW) management and contaminated site clean-up

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Abstract: This chapter presents the French experiences of contaminated site clean-up and remediation. Radioactive waste management in France is discussed in general terms including the classification of waste. The history of the French waste management organization including site remediation is then discussed, highlighting difficulties encountered and lessons learned.

Key words: site remediation, classification of waste, waste management, waste management organization, orphan polluted sites, conventional risk, radiological risk.

15.1 Introduction

To understand the subject of overall radioactive waste (RAW) management in France, it is important to first describe the sources of waste and the associated classification system, knowing that the latter also has a rationale linked to repository availability. It is then important to describe the waste management organization, its history and its current status. The subject of site remediation can then be addressed, first discussing the waste management organizations, past and present, before describing the site remediation activities.

15.2 Sources, types and classification of wastes

15.2.1 Waste sources and categories

The various types of RAW are classified according to the half-lives and radioactivity levels of the main radionuclides they contain, to their physical and chemical characteristics, as well as to their origins. Half-lives are divided into very-short (less than 100 days), short (between 100 days and 31 years) and long (over 31 years).

In France, there are six major waste categories depending on their radioactive content (activity level and half-life), as follows:

- High-level waste (HLW) consists mainly of vitrified-waste packages in the form of stainless-steel containers, which contain the vast majority of radionuclides, whether in the form of fission products or of minor actinides. Radionuclides contained in spent fuel are separated from plutonium and uranium during fuel reprocessing at the La Hague plant. The activity level of vitrified waste is on the order of several billions of Becquerels per gram.
- Long-lived intermediate-level waste (LL-ILW) originates mostly from the reprocessing of spent fuel and consists of structural residues from nuclear fuel (i.e., hulls (sheath sections) and ends, which were conditioned initially into cemented waste packages, but are now compacted into stainless-steel containers). It also includes technological waste (e.g., used tools, equipment, etc.) and residues resulting from the processing of effluents, such as bituminized sludge. The activity of these residues ranges between 1 million and 1 billion Becquerels per gram. There is either no or a negligible heat release.
- Long-lived low-level waste (LL-LLW) consists mainly of graphite and radium-bearing waste. The activity of graphite waste lies between 10,000 and 100,000 Becquerels per gram. Its long-term activity arises from long-lived beta-emitting radionuclides. Radium-bearing waste contains long-lived alpha-emitting radionuclides and their activity lies between a few tens to a few thousands of Becquerels per gram.
- Short-lived low- and intermediate-level waste (SL-LILW) results mainly from the operation and dismantling of nuclear power plants (NPP), fuel cycle facilities and research establishments, as well as, for a small amount, from activities relating to biological and academic studies. Most residues in this category were disposed of in a surface facility at the Centre de la Manche disposal facility (CSM) up until 1994 and at Centre de l'Aube disposal facility for LILW (CSFMA) since 1992.
- Very-low-level waste (VLLW) is mostly from the operation, maintenance and dismantling of NPPs, fuel cycle facilities and research establishments. Its activity level is generally lower than 100 Becquerels per gram. All residues of this category are disposed of at the Centre de l'Aube disposal facility for VLLW (CSTFA).
- Very-short-lived waste includes residues that result notably from medical uses.

For practical purposes, the acronyms listed in Table 15.1 are often used.

15.2.2 Classification of waste in France and management of different categories

Table 15.2 presents each waste category along with the current identified long-term management solution. For some categories, the corresponding

Table 15.1 Acronyms used for the different waste categories

Acronyms	Designation	French acronyms
HLW	High level waste	HAVL
LL-ILW	Long-lived intermediate level waste	MA-VC
LL-LLW	Long-lived low-level waste	FA-VC
SL-LILW	Short-lived low- and intermediate-level	FA/MA-VC
VLLW	Very-low-level waste	TFA

Table 15.2 Classification of waste

<table>
<tr><th>Half-life / Activity</th><th>Very short half-life
(<100 days)</th><th>Short half-life
(≤31 years)</th><th>Long half-life
(>31 years)</th></tr>
<tr><td>Very low level (VLW)</td><td rowspan="4">Management by radioactive decay</td><td colspan="2">Surface disposal (CSTFA)</td></tr>
<tr><td>Low level (LLW)</td><td rowspan="2">Surface disposal (CSFMA)</td><td>Systems under study pursuant to Article 3 of the 2006 Planning Act</td></tr>
<tr><td>Intermediate level (ILW)</td><td>Systems under study pursuant to Article 3 of the 2006 Planning Act</td></tr>
<tr><td>High level (HLW)</td><td colspan="2">Systems under study pursuant to Article 3 of the 2006 Planning Act</td></tr>
</table>

long-term management solution is still under study and this issue is addressed in the 2013 National Plan for the Management of Radioactive Materials and Waste ('Plan National pour la Gestion des Matières et Déchets Radioactifs' or PNGMDR), which is a three yearly plan stating, for all radioactive materials and waste in France, the chosen long-term management option, either operational or being researched.

There is no simple and single criterion to classify RAW. There is no overall activity level, for instance, to determine whether a given residue belongs to the SL-LILW category. It is necessary to examine the radioactivity of the different radionuclides present in the waste in order to rank it according to the classification. More particularly, in order to be considered as SL-LILW, the specific activity of each radionuclide in the waste must be lower than the prescribed thresholds in the waste acceptance specifications for the SL-LILW disposal facility ('Centre de Stockage de déchets de Faible et Moyenne Activité' or CSFMA; see Fig. 15.1 for the different facilities

15.1 Map of French facilities managed by ANDRA.

managed by ANDRA). In that category, the activity of long-lived radionuclides is particularly limited.

However, it is possible to indicate a range of specific activities within which each waste category generally belongs. It may be that a specific waste pertaining to one of the above-mentioned categories is not acceptable within the corresponding management system due to other chemical, physical or other characteristics. Such is the case for residues containing significant quantities of tritium (a radionuclide that is difficult to confine or retain) or of sealed sources for medical uses.

A special case also concerns the waste generated by uranium enrichment facilities and fabrication plants of nuclear fuel containing uranium oxide. Their waste residues contain uranium and are compatible with the acceptance criteria of the CSFMA or, if their activity is very low, with those of the VLLW repository ('Centre de Stockage de déchets de Trés Faible Activité', or CSTFA). In the first case, the waste is disposed of at the CSFMA and, by convention, registered as SL-LILW, notably in the national inventory. In the second case, the waste is disposed of at the CSTFA and included in the VLLW category.

15.3 Radioactive waste (RAW) management strategies: history and developments

15.3.1 1969–1991

The history of radioactive waste management began long before the creation of ANDRA. In France, the first radioactive waste was produced in the 1930s, especially in hospitals that used sources of radium to treat cancer.

With the creation of the Atomic Energy Commission (CEA), after the Second World War, nuclear research centres began to generate radioactive waste in larger quantities. There was no organized disposal for this waste, although rules based on radiotoxicity levels already existed. The chosen option was to condition the waste safely, and to store it on the CEA sites.

In the 1950s and 1960s, with the appearance of France's first NPP, the development of research and manufacture of nuclear weapons, significant quantities of radioactive waste began to accumulate. In 1967 and 1969, France took part in two international experimental campaigns, organized by the OECD/NEA, on waste immersion before giving this practice up as unsatisfactory.

Meanwhile, the CEA decided to set up next to its fuel reprocessing plant at La Hague a centre to dispose of short-lived low- and intermediate-level wastes. The Centre de la Manche started operations in January 1969.

The creation of ANDRA

In 1974, due to the oil crisis that followed the Yom Kippur War, France decided to develop a very large nuclear industry, including a number of NPPs and units to recycle spent fuel. The initiation of this significant programme had the effect of greatly increasing the volume of all categories of RAW: that of high-level and long-lived waste, coming from the recycling of the spent fuel, as well as that of short-lived low- and intermediate-level waste.

To address this situation, the government asked the CEA to create within its ranks an organization to take responsibility for managing all this waste. ANDRA was created inside the CEA in 1979.

The management of the Centre de la Manche disposal facility

The first task assigned to ANDRA was to operate the surface disposal of SL-LILW that had been created in 1969 at the Centre de la Manche. It also laid down some rules to secure and streamline disposal of waste. For example, the waste had to be packaged in standard packages. In addition,

ANDRA built a collection system to monitor and control the water coming out of the disposal facility, which allowed the impact of the centre on its environment to be monitored.

A new disposal centre in the Aube district

From 1984, ANDRA began looking for a new site for a disposal facility to replace the Centre de la Manche. Geological studies were undertaken in different 'Departments' (the 'Department' is the main political and administrative subdivision in France). In 1984 and 1985, more than 500 boreholes were drilled in the Aube Department to select a specific location. At this time, ANDRA perfected the technique of using a multi-barrier system consisting of the package, the engineered barrier and the geology to dispose of the waste. Meanwhile, local consultation was carried out through the organization of several visits and meetings with local stakeholders. On 22 July 1987, the Prime Minister signed the Declaration of public interest: the new disposal facility for SL-LILW, the CSFMA, was located in the Aube Department, near the village of Soulaines-Dhuys.

Initial research for a deep geological disposal facility

Since the late 1960s, HLW and LL-ILW, mainly from French nuclear fuel processing, has been stored in the plants at La Hague and Marcoule pending a final disposal option. The proposed solution was to dispose of them in deep geological formations – stable for millions of years – the only ones likely to contain the waste effectively over these timescales. Therefore, in 1982 ANDRA began to collaborate with countries that are already carrying out detailed studies of various rocks for underground disposal:

- in Mol, Belgium, in clay,
- in Asse, Germany, in a former salt mine,
- at Grimsel, Switzerland, in granite.

In 1987, after documentary studies, ANDRA tasked its geologists with conducting investigations at four sites in France where the geology was deemed favourable to the installation of underground laboratories to study the feasibility of a deep repository:

- in the Deux Sèvres Department (granite)
- in Maine et Loire Department (shale)
- in the Ain Department (saline formations)
- in the Aisne Department (clay).

However, public protests in these territories meant that after three years, the Prime Minister, Michel Rocard, eager to break the deadlock on a very

important issue for the development of the nuclear industry, announced a one-year moratorium for all these projects.

The 1991 Act: the independence of the Agency

In December 1991, the National Assembly passed a law that gave ANDRA a new status making it independent of the CEA (see ANDRA, 1991). This law regulated in detail the feasibility study related to disposal of waste in deep geological rocks. The Agency had 15 years to complete this study. Christian Bataille (Member of Parliament for the Nord Department) was responsible of a mediation mission to seek local volunteers for the hosting on their territory of an underground laboratory.

15.3.2 1992–1999

1992–1994: ANDRA pursues its mission

In January 1992, the disposal facility CSFMA received its first waste package for disposal in concrete vaults. Also in 1992, ANDRA began to develop a specific management solution for waste coming from outside the nuclear power industry, particularly that from hospitals and used for medical training. This complex development work took nearly 10 years. Finally, in the disposal facility of the Centre de la Manche, the last package arrived on 30 June 1994, after 35 years of operations. The implementation of the waterproof cover continued, in view of the transition to the monitoring phase in 2003.

1994–1996: ANDRA conducts further investigations to establish a geological underground laboratory

With the success of the mediation mission assigned to Christian Bataille, whose objective was to conduct preliminary consultations to propose to the government favourable sites for the implementation of underground laboratories, teams from ANDRA returned to fieldwork in 1994. They performed geological investigations in four Departments:

- Gard (clay),
- Vienna (granite),
- the Meuse (clay),
- the Haute-Marne (clay).

Through seismic campaigns and core drilling, the geological layers that could accommodate a laboratory were determined. This was done under very different conditions from those existing prior to the moratorium. First, a law now regulated the action of ANDRA. Second, around the sites, local elected officials supported the Agency. There was still some opposition,

demonstrations, and some malicious acts, but the work of ANDRA was not hindered.

In 1996, projects for the Meuse and Haute-Marne Departments were combined in a single site located in the town of Bure. ANDRA then filed three applications for installation of underground laboratories. However, in 1997, political difficulties prevented any decision. In the new French government of Lionel Jospin, the underground laboratory project was no longer unanimously accepted.

1998: ANDRA now has permission to establish a laboratory in clay

ANDRA's research on the feasibility of an underground facility did not stop. From 1996 to 1998 the Agency joined the 'Mont Terri' Swiss project in the Jura, where researchers were using the viewing gallery of a motorway tunnel to conduct experiments on a clay layer with qualities similar to those of the clay at Bure. ANDRA also continued to fund research in many university laboratories.

In August 1998, a large European anti-nuclear gathering took place at Bure. That day, the mayors of a dozen neighbouring municipalities installed signs 'Yes to the lab' to the fronts of their town halls. In December 1998, a political compromise was found and the government announced its decision:

- future storage must be reversible.
- the site of the Gard was discarded.
- the research on the granite site of the Vienne Department was considered inconclusive, but ANDRA should nevertheless continue to study the rock.

Finally, the Meuse/Haute-Marne site was chosen to implement an underground laboratory: more than 10 years after its first research (1987), ANDRA had the permission to create a laboratory in clay.

1999: a new storage centre in Aube

Discussions were initiated with stakeholders (elected representatives and associations) for the opening of a new disposal facility for VLLW (CSTFA) near the CSFMA. Studies began to determine the most favourable area from a geological point of view.

15.3.3 2000–2006

In early 2000, the laboratory is being built

The digging of a shaft is in itself a laboratory: a few metres were dug, and then the site was assigned to geologists for them to study the rock for a few

hours before the shaft digging resumed. Unfortunately, in May 2002, following a fatal accident, the site was closed for almost a year.

2001–2003: creation of the new repository for VLLW

In 2001, a first public hearing on the utility of creating a centre for low-level waste and clearing land for the future centre was held. Then, a second hearing was held in 2002 concerning the authorization to operate and the building permit request. On 9 August 2002, the building permit was issued by the Department 'prefet', and, on 26 June 2003, he also issued the operating permit for the centre.

Thus, with the opening in the town of Morvilliers of the CSTFA, the first centre of its kind in the world, ANDRA found a solution for managing waste from the decommissioning of nuclear facilities. Waste packages were stored in vaults dug in the clay, and protected from the elements by a large removable roof before being permanently covered with a layer of clay several feet thick.

2004: completion of the underground laboratory

In November 2004, in the underground laboratory of Meuse/Haute-Marne, ANDRA reached at 445 m depth, the depth of the clay layer to be studied. The first gallery was dug to install a series of experimental devices. Then the digging of the shafts continued, and at 490 metres, new galleries were dug horizontally and other experiments undertaken. The data collected fully confirmed the results of laboratory research and tests made at Mont Terri in Switzerland.

2005: submission of Dossier

A few months later, ANDRA released the Dossier 2005 (ANDRA, 2005). In 10,000 pages, it compiled 15 years of research. It concluded that the clay layer of the Meuse/Haute-Marne was perfectly suitable for receiving a disposal facility for high- and intermediate-level long-lived radioactive waste. This dossier also contained a description on the interest in granitic formations for geological disposal. A milestone had been reached for the management of radioactive waste in France.

15.3.4 2006–2010

2006: new legislation, new missions

The Act of 28 June 2006 reinforced ANDRA's mission of design and operation of RAW disposal and also requested that ANDRA have a 'public service' mission (ANDRA, 2006):

- The Agency must develop, in the clay layer at 500 m depth in the Meuse and Haute-Marne districts, a reversible deep repository for high level and intermediate long-lived waste.
- It must also seek a solution for low-level long-lived waste, both radium-bearing and graphite.
- The 'public service' missions were two-fold:
  - Remediation of former radioactively contaminated so-called orphan sites (i.e., those for which there is no responsible body) is managed in a more sustainable manner with the creation, as from 2007, of the National Commission for Assistance in the Radioactive Area ('Commission Nationale des Aides dans le domaine Radioactif', or CNAR), decided by the Board of ANDRA.
  - The management of radioactive waste obtained from individuals (e.g., alarms with radio luminescent needles, radium fountains – radium was believed in the past to have therapeutic virtues and such fountains for radium distribution can still be found in French households, etc.) was addressed with the launch in late 2008 of a campaign (involving the 36,000 mayors of France, the departmental services for fire and rescue, the waste treatment entities, among others) to identify and remove such radioactive objects from homes and manage them safely through storage or disposal.

2008: looking for management solutions for LL-LLW

In June 2008, having identified 3,115 boroughs with a potentially favourable geology for the repository for long-lived low-level waste, ANDRA sent a call for volunteers through an information document to the mayors of the municipalities concerned. At the end of 2008, more than 40 municipalities declared themselves candidates to analyze the opportunity of such a repository. In June 2009, based on an analysis conducted by ANDRA, the government chose two of them (Auxon and Pars-lès-Chavanges in the Aube Department) in which to conduct thorough geological and environmental investigations. However, under pressure from opponents, both municipalities withdrew from the project in July and August 2009. In June 2010, in the National Plan for the Management of Materials and Radioactive Waste (PNGMDR, 2010), the State set new guidelines for the project: based on further studies on knowledge, treatment and conditioning of LL-LLW, ANDRA must submit to the government (no later than 2012) a report outlining possible management scenarios for these wastes.

2009–2010: Birth of the industrial centre for geological disposal (CIGEO)

Next to the underground laboratory, a technology centre was constructed in 2009 in which are now displayed the prototypes of objects and machines

that demonstrate what could be implemented in the disposal facility. In late 2009, ANDRA produced a 'dossier' (ANDRA, 2009) giving the status on the development of the planned repository. For the study of the location of underground facilities of the repository, it proposed an area of 30 km^2 called the Area of Interest for Detailed Investigation ('Zone d'Intérêt de Recherche Approfondie', or ZIRA), that was the result, apart from scientific considerations, of a dialogue with local stakeholders. In March 2010, ANDRA was authorized to conduct detailed geological investigations in this area. The project Industrial Centre of Geological Disposal ('Centre Industriel de stockage GEOlogique', or CIGEO) was launched.

15.4 Contaminated site clean-up experience

15.4.1 Organizational set up until 2006

The contaminated site clean-up mission was initiated in the early 1990s. The first site to be operated on was the so-called 'Bayard' site, linked to the watch-making industry. Between 1995 and the 2006 milestone, operations on contaminated sites were handled on an ad hoc basis with the different sites being worked case by case, and with specific project/coordination structures.

In fact, ANDRA intervened on the basis of the circular of 16 May 1997 and benefited from two mechanisms for financing:

- one dedicated to orphan polluted sites known as the agreement on orphan polluted sites('Sites Pollués Orphelins', or SPO),
- one fund concerning radium-bearing object collection, when additional resources other than ANDRA's were required.

The SPO agreement was based on financial sponsorship by the waste producers CEA, EDF and Areva. The decisions were taken by a programme committee and based on reporting by ANDRA. The main objective was the safe management of the collected waste. This agreement was discontinued in 2005 and was not renewed. The fund concerning radium-bearing object collection was established applying the fund management mechanism used within the environment agency. Decisions were taken by a committee chaired by the Ministry of Industry. The radium-bearing object collection concerned only the radium objects dating from the interwar period.

These two mechanisms of financing suffered from the limitation of their objectives and of their temporary character.

15.4.2 Current organization (since 2006)

The Legislator decided in 2006 to endow the Agency with a clear framework (executive) for intervention on contaminated sites.

Law number 2006-739 of 28 June 2006 (ANDRA, 2006) relating to the sustainable management of radioactive materials and waste specifies the public service missions of ANDRA by setting three objectives:

- Determining and publicizing the national inventory of radioactive materials and waste.
- Management of certain wastes from the general public, in particular when the public, totally foreign to any use of radioactivity, become holders of radioactive objects (by inheritance, for example) sometimes unaware of the radioactive nature of the objects which they hold (e.g., radium objects),
- The remediation of former radioactively contaminated (orphan) sites and the management of the waste generated.

The law included the principle of a State subsidy contributing to financing the missions of general interest entrusted to the Agency.

The financing mechanisms described above in Section 15.4.1 is replaced by an annual public subsidy securing stable financing of the operations and thus allowing the programming of multi-annual intervention according to the site prioritization. The public subsidy also allows total financing of the works, and makes the situation of certain private individuals easier (e.g., when they cannot finance, or only in part, the rehabilitation works to their property).

The public subsidy also allows financing of storage of polluted soils at the remediation sites and the storage of radium-bearing objects that require, in the absence of a definitive solution (disposal of LL-LLW – see Section 15.2.2), to be stored on dedicated sites (located on the CEA sites in Saclay and Cadarache). Of course, in parallel, ANDRA strives to minimize the volume of polluted soils coming from remediation sites.

To manage this work on remediation, a new department was created in ANDRA in January 2007 (within the Industrial Direction). Its role is to lead and coordinate the Agency's work on the remediation mission. Deliberations on the decision-making structure for the use of the State subsidy was the object of extensive work from 2006 to the beginning of 2007 with participants from the Ministry of Industry and from the Safety Authority. This work resulted in the creation of a National Commission for assistance in the Radioactive Area (CNAR) which expresses an opinion on the use of this public subsidy, on the allocation priorities for the funds, on the strategies of treatment of the polluted sites and on the questions of doctrine regarding the waste.

The functioning of this committee is similar to that of other structures with similar missions in the governmental sphere (such as those for environmental remediation). The CNAR, chaired by the Chief Executive Officer

of ANDRA, includes representatives of the authorities (Safety Authority, the appropriate ministries, technical public institutions such as the French Technical Support Organization ('Institut de Radiprotection et de Sûreté Nucléaire', or IRSN), NGOs: two environmental protection associations, elected representatives) and two qualified persons (a representative of a public institution and a specialist in remediation).

The CNAR was created by deliberation of ANDRA's Board of Directors in April 2007. It met twice in July and September 2007 and immediately began to discuss operational issues.

In 2012, the structure remains active, unchanged, and is handling the remediation mission.

15.4.3 Analysis of context of the site remediation

Overview

The sites for remediation are:

- Old industrial sites:
  - from the radium industry which flourished in the interwar period
  - from the production of objects for medical or daily use
  - from the production or the usage of radium paints for watches, clocks, military instruments
  - from the production or the usage of tritium-based paints from 1960
  - from the extraction of caesium from monazite
  - from tracer fabrication (more recently)
  - where rehabilitation is pending
- Old laboratories dealing with radioactivity.

Analysis of the remediated sites

The sites of radioactive pollution for which the Agency intervenes are mainly former (ancient) installations having used or made radium or thorium and for most of which the owner can no longer be traced.

In two cases out of three, these sites have their origins in the prosperous interwar radium industry that involved factories for the extraction of radiferous ore (Gif-sur-Yvette, Nogent-sur-Marne, Saint Denis island) or workshops using radium products such as radioluminescent paints (e.g., in the watch-making industry or for clock manufacturing). The sites of the watch-making industry may also have tritium pollution following the replacement of radium by tritium in the 1960s. Other contaminated sites arise from rare earth extraction (thorium) and, more recently, manufacture of tracer molecules (tritium and carbon-14, in particular).

The age of these sites frequently raises the problem of records of these activities, and lack of description of what has been allowed in the past (such as inappropriate use of sites from the point of view of radioprotection; see Fig. 15.2). Indeed, some of these sites are in densely housed areas, such as in Gif-sur-Yvette, and others can be found on the site of sensitive activities such as day nurseries, or schools. Other sites are in a safe state and await requalification. The problem of lack of records arises when intervention is envisaged. Mostly archives are unavailable, the owners of the site have often disappeared or died or refuse to communicate their knowledge. The interventions thus have to base themselves on very precise mappings and soundings of grounds to know the state of pollution and, if necessary, analysis of

(a)

(b)

15.2 (a) and (b) Examples of abandoned contaminated industrial sites.

subterranean waters. This stage of characterization is long and expensive, but is essential.

Naturally any zoning of the waste on these sites is impossible. Packages of waste have to be characterized on-site, taking into account the existing radioelements, the nature of the materials and the planned modes of conditioning. This can add technical difficulties in the case of lack of space or of raised levels of radiological protection. Most of the time the waste recovered is mainly VLLW and to a lesser extent, radiferous. Some cases are made even more complex, such as in the case of mixed chemical and radioactive waste, for which the solutions must be studied separately.

The great majority of sites are in the Paris region and in densely populated urban zones, so that the site cannot be reopened for industrial use. The question of the requalification of the site is directly linked to the definition of the rehabilitation objectives. This definition is the topic of discussion with the local safety authority. Also, the location of these sites in urban zones raises difficulties linked to the creation of a decommissioning construction site. This requires frequent contact with local authorities to find satisfactory solutions to the required limitation of hazards to the local residents. The characteristics and problems of these sites are closer to those of chemically polluted sites than to those of decommissioning in a nuclear environment.

ANDRA's missions in this domain are similar to those of the environmental agency in the field of chemical pollution, with comparable intervention mechanisms (although the number of sites is much smaller for ANDRA):

- There are about 20 radioactive contaminated sites for ANDRA and approximately 150 chemically polluted sites for the environmental agency requiring immediate intervention.
- In total there are about 50 sites of radioactive pollution against approximately 4,000 sites of chemical pollution.

Operationally, ANDRA intervenes on sites at the request of public authorities. This requisition generally takes the shape of an order from the Department prefet based on the legislation for industrial landfills. The prefet makes this order after authorization from the Minister for Ecology. The interventions are made in close collaboration with the local administration concerned in the areas of safety, worker safety and industrial safety.

15.5 Problematic cases and lessons learned

15.5.1 Difficulties encountered

Lack of accurate historical records requires expensive and accurate site characterization such as:

- cartography, underground water sampling, earth probing, ecological sampling sometimes on sites in an urban environment generally (see Fig. 15.3),
- setting up the works and storing waste packages in small areas,
- waste package storage on the urban site before evacuation,
- nuisances during operations (noise, truck traffic) for neighbourhood, relations with private owners,
- health and patrimony concerns.

Also there is currently no disposal for LL-LLW in France (such as Ra-226 with levels above 10 Bq/g) and this implies:

- use of interim storage,
- cost of interim storage and limited capacities.

Those difficulties require flexibility in the remediation project management.

Another difficulty concerns local stakeholders and for this a helpful initiative is that of the local communication initiative (organizing public meetings, for example).

15.3 On-going decontamination work inside a private house.

15.5.2 Lessons learned

Absence of those responsible

Polluted sites are predominantly from the radium mining industry. The industry died out in the 1920s after a boom period in the wake of the work of Marie Curie and medical applications implemented during the First World War. Thus the last radium extraction site stopped in 1928 (in the town of Nogent-sur-Marne). It is thus unrealistic to look for any responsible body still in place (there is now a 30-year proscription applicable to industrial activities following the decision of the State Council on the 'Allusuissse' issue). The situations ANDRA inherits are therefore situations from the national industrial heritage, to be managed as best as possible.

Sites are almost always reused for other purposes: innocent holders

The logical consequence of the age of the sites and their predominantly urban characteristic (the Paris area is the historic cradle of the radium industry) is that sites have usually been reused for other purposes, including for housing. As a result, the current owners cannot be treated as responsible for the pollution that affects them. These occupied sites are in fact considered as 'assimilated to orphans' sites, although they have an owner present. This question raises starkly the two questions of health during the works and inheritance aspects for the management of these sites, aspects that have been neglected in the past.

ANDRA personnel are used to (and are chosen for their ability to) incorporate these aspects in their delicate human relations with residents. However, any difficulties or particular relational situations (and issues potentially difficult to live with) should immediately be shared with management to find adequate answers so that no added burden is put on the personnel involved. In any case, it is important to remain aware of the psychological and social dimension of the remediation of these sites.

Total clean-up is mostly illusory

If might be intellectually satisfying to seek total clean-up, but experience shows that this goal is often illusory and generates unnecessary costs. Indeed, what does total clean-up mean? Should we seek to return to the background noise level, regardless of the cost incurred? The house in Gif-sur-Yvette shows that even having reached a dose rate for Ra-226 in the range of background noise (0.1 mSv/h) in the home, radon levels remain significant and close to the pseudo-limit of 400 Bq/m^3. Substantial resources have been committed without the possibility of cleaning the house

completely, simply because working on the house itself without addressing the surrounding land amounted to moving the pollution limit without eliminating it (radon, in its migration in the ground, ignores administrative boundaries). Total clean-up is only possible for localized pollution. Even then, it is still necessary to agree on a target value for pollution control, and therefore on the residual contamination that is left behind, which *de facto* contradicts the idea of total clean-up.

Similarly, on an industrial site, considerable sums were spent to treat the site, and thus produced contaminated soil now stored at the CEA (the Cadarache site). The cost of disposal of the soil is assessed at over € 2 million even though the original site is still not completely cleaned up. The pursuit of an illusory goal of total clean-up has led to considerable – and probably unjustified – expense without the goal being reached.

In its communications with the media, ANDRA must refrain from using a term as misleading and meaningless as the 'total clean-up'.

An activity marked more by conventional than radiation risk

Cutting, working from heights, working on building structures, the presence of asbestos, the use of electrical systems, work on sites that used chemicals, work on sites often unknown from a chemical point of view mean that actions on contaminated sites present conventional risks, much more dangerous in terms of severity, risk of occurrence and kinetics, than the radiological ones. ANDRA's agents are therefore sometimes asked to balance a number of conflicting risks between the need to control conventional risk and radiological hazards. It is therefore necessary, in preparing a site, to reconcile the requirements in terms of both conventional and radiological risk control with the security engineer who will be able, after technical dialogue, to decide how to act and to best manage these risks via a suitable protocol. In any case, priority will be given to systematic conventional risk management except in exceptional cases. Also on the sites of major pollution, the use of a safety coordinator will most often be used.

Know where to intervene

Experience has shown clearly that the initial characterization of sites is paramount. All past projects that have been engaged on the basis of inadequate initial characterization have ultimately led to hard to overcome technical difficulties and additional costs far higher than the savings attained on the initial characterization of the site. No concessions should be made on the initial characterization of the site. ANDRA's agents should also not

intervene for site remediation on a site that has not been sufficiently characterized.

Know when to assist

Experience also shows that many operations rely on trades from different core businesses than that of the 'remediator'. In particular, sites often rely on experts for asbestos removal, demolition of buildings, work on hydrology, management of conventional toxic waste, management of radon in buildings, the maintenance of building structure, etc. ANDRA's agents have a duty to draw on the external expertise needed to better adress the risks that are poorly controlled or not controlled at all inside ANDRA. Again, any initial savings on these aspects can cost dearly later.

An activity that is a waste generator

ANDRA's agents may be producers of waste in their own right: any site remediation results in the generation of waste. Therefore agents must, before the remediation work, measure and characterize the waste from remediation projects. There again, experience shows that taking these issues into account upstream minimizes delays in administrative treatment and hence the storage time on the polluted site for the waste packages awaiting decision. This delay can be tricky if the owner is a private individual.

Similarly, for certain categories of waste, the agent may need to discuss with those responsible for a disposal site (current or planned), defining upstream the best conditions for characterization, packaging and management of waste arising from remediation sites. In any case, ANDRA as a waste generator must be exemplary in terms of waste and therefore how to take care of it.

15.6 Future trends

Two major areas of progress for this activity are:

- Technology, and site characterization is already calling on the use of sophisticated equipment such as aerial or mobile means. Aerial means are useful at sites where the true extent of the contamination is unknown, and therefore large areas have to be investigated.
- Stakeholder involvement and ANDRA is committed to working with those that are involved in this activity, such as private owners whose homes have been found to be contaminated or the residents around a contaminated industrial site.

Furthermore, and regarding the waste arising from the decommissioning of the fleet of French reactors, the site remediation issues are managed by the waste producers and ANDRA is only responsible for the disposal of the corresponding waste, either in one of its operational sites or in future projected sites.

15.7 Sources of further information

A lot of information on the site remediation activity in France is available from ANDRA's website: www.andra.fr. Other information is also available from: www.asn.fr (Safety Authority website) and www.irsn.fr (Technical Support Organisation website).

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16

England and Wales: experience of radioactive waste (RAW) management and contaminated site clean-up

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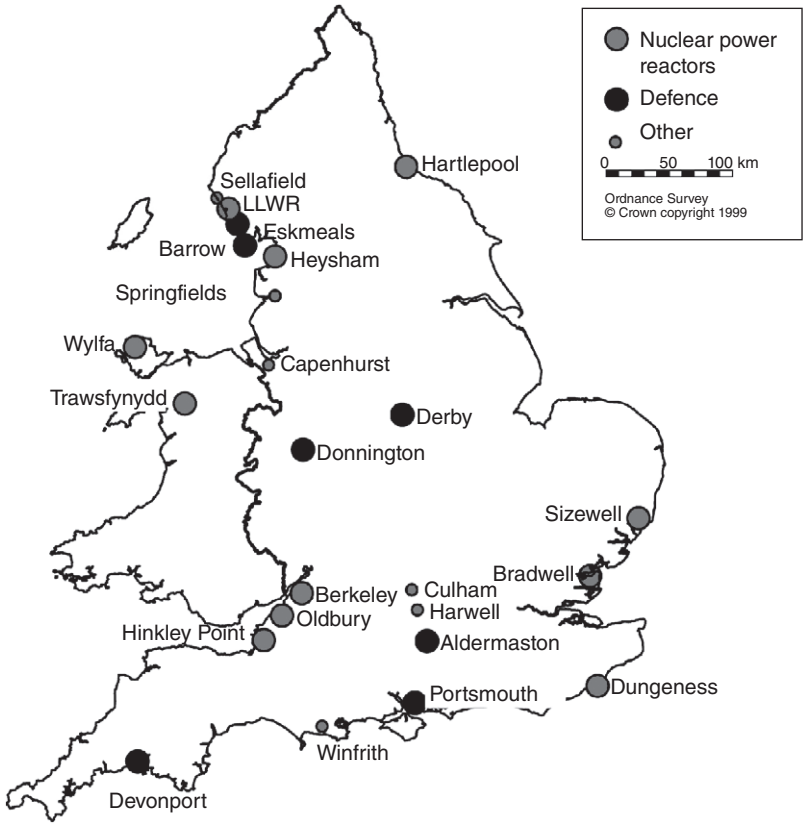
Abstract: The United Kingdom has a long history of nuclear development. Waste management principles and strategies have evolved over this period, together with technical developments allowing modified waste management regimes. High and intermediate level wastes reflect both current arisings and legacy wastes, with disposal and storage options being explored and implemented. In recent years, low level waste strategy has been further clarified to expand options available for safe and cost-effective disposal. Challenges with respect to contaminated land and delicensing of decommissioning sites are recognised. Devolution of waste management responsibilities within the UK is leading to some divergence in national policies, particularly with respect to higher activity wastes.

Key words: Nuclear Decommissioning Agency (NDA), Magnox, advanced gas-cooled reactor (AGR), reprocessing, waste policy.

16.1 Introduction

The United Kingdom has a long history of nuclear development, which, for convenience, can be traced from the post-war weapons programme and, later, the civil use of nuclear power. Research and production sites in England at Harwell (Oxfordshire), Sellafield (Cumbria), Springfields (Lancashire) and Capenhurst (Cheshire) were established in the 1940s and, in Scotland, the Dounreay site (Caithness) followed in 1954, initially to develop the fast breeder reactor.

The UK's first commercial nuclear power reactor began operating in 1956 and, at its peak in 1997, 26% of the nation's electricity was generated from nuclear power. Nuclear reprocessing facilities were also built to deal with the increasing demand from both military and civil programmes. Since then a number of stations have been closed, and others are scheduled to follow over the next decade. Of the currently operating stations, lifetime extensions may be granted for some sites, allowing for continued generation until



16.1 Map of all major nuclear installations in England and Wales. Coastline map reproduced from Ordnance Survey map data by permission of the Ordnance Survey © Crown copyright 1999.

replacement generating sources become available. Locations of all major nuclear licensed sites in England and Wales are presented in Fig. 16.1.

This account of radioactive waste (RAW) management in England and Wales is oriented towards the strategic and environmental issues arising from the management of RAW from the nuclear industry. It also addresses the structure of the nuclear industry and the sources, types and classification of RAW.

Approximately one million m^3 of solid RAW has been disposed of in the UK to date (NDA and DECC, 2011). Current wastes identified, plus projected wastes over the next century or so, amount to around 4.7 million m^3 in the UK. About 97% (4.6 million m^3) of the total volume of RAW anticipated has already been produced. Some has been processed, and is being held in stores, but most is contained within existing nuclear facilities, including reprocessing plants and nuclear reactors, and will not be processed until

these are shut down and dismantled. This waste is the legacy of past and current civil and military nuclear programmes. About 3% (150,000 m^3) of the radioactive waste total has yet to be produced. This waste is that forecast from the future planned operations of the existing nuclear power industry, from ongoing defence programmes and from the continued use of radioactivity for medical and industrial purposes.

Current and projected radioactive waste volumes for England and Wales are summarised in Table 16.1 (NDA and DECC, 2011).

16.2 Structure of the UK nuclear industry

Prior to the break-up and privatisation of the electricity generation industry in the 1980s and 1990s, the operators of nuclear installations were primarily government-owned organisations. More recently, the UK government has given the go-ahead for a new generation of nuclear power stations to be built. Potential sites have been identified across England and Wales. However, the devolved Scottish government has no current plans for new nuclear power stations. A divergence in approaches to waste management between Scotland and the remainder of the UK has also been confirmed (Defra *et al*., 2008; Scottish Government, 2011). Northern Ireland currently has no nuclear power stations and no identified sites for potential new build, although there is no policy restricting the development of nuclear power in Northern Ireland.

Table 16.1 Current and projected radioactive waste in England and Wales

Waste type	Volume (cubic metres)		
	Stocks at 1 April 2010	Estimated future arisings	Lifetime total once all wastes are packaged
England			
HLW	1,620	–601[a]	1,330
ILW	83,200	164,000	424,000
LLW		35,000	4,040,000
		4,010,000	
Wales			
HLW	0	0	0
ILW	3,070	10,800	22,300
LLW	539	106,000	132,000

[a]Future arisings of HLW in England have a negative volume. This is because Sellafield has reported future arisings of HLW to show that the volume of accumulated waste (liquid plus vitrified product) will fall as liquid waste existing at 1 April 2010 and forecast in the future is conditioned to a vitrified product. No HLW is managed in Wales.

Table 16.2 Status of reactors in England and Wales, October 2011

Power station	Type	Net MWe	Construction started	Connected to grid[a]	Status
Oldbury	Magnox	434	1962	1967	Closed 2012
Wylfa	Magnox	980	1963	1971	Operational
Dungeness B	AGR	1,110	1965	1983	Operational
Hinkley Point B	AGR	1,220	1967	1976	Operational
Hartlepool	AGR	1,210	1968	1983	Operational
Heysham 1	AGR	1,150	1970	1983	Operational
Heysham 2	AGR	1,250	1980	1988	Operational
Sizewell B	PWR	1,188	1988	1995	Operational
Calder Hall	Magnox	200	1953	1956	Closed 2003
Berkeley	Magnox	276	1957	1962	Closed 1989
Bradwell	Magnox	246	1957	1962	Closed 2002
Hinkley Point A	Magnox	470	1957	1965	Closed 2000
Trawsfynydd	Magnox	390	1959	1965	Closed 1991
Dungeness A	Magnox	450	1960	1965	Closed 2006
Sizewell A	Magnox	420	1961	1966	Closed 2006

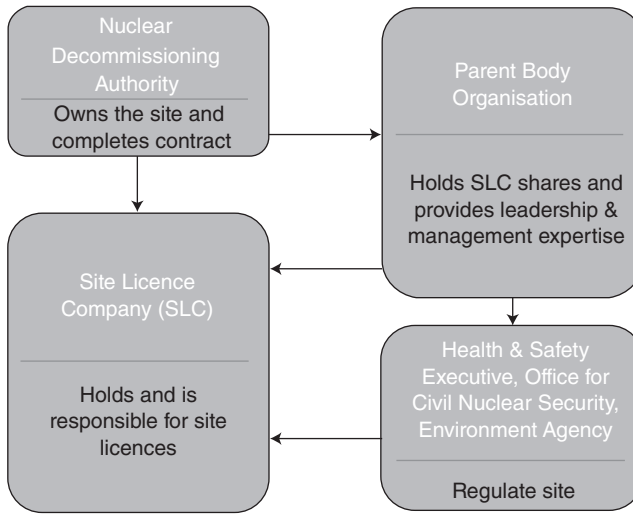
[a]For sites with multiple reactors, the date of connection to the grid represents connection of the first reactor unit (e.g., Calder Hall had a four reactor design. Reactor 1 was connected in 1956; reactor 4 was connected in 1959).

RAW management in Scotland is considered in Chapter 17, but there is considerable overlap with England and Wales, and for much of the earlier history of nuclear developments, a UK-wide policy was applied.

As the older stations and other facilities have closed, a significant liability has accumulated, much of which has been retained in the public sector as privatisation of facilities reaching the end of their working lives was not practicable. The current status of reactors in England and Wales is summarised in Table 16.2.

A number of research and development reactors also produced some power for the grid, including two Winfrith reactors, two Dounreay fast reactors, and the prototype Windscale advanced gas-cooled reactor.

The Nuclear Decommissioning Authority (NDA) was established in 2005 to take on the role of addressing the nuclear legacy from these older sites in a planned and focused manner. The NDA is responsible for the largest current decommissioning and waste management liabilities in the UK, overseeing the continued operation, decommissioning and site clean-up at 19 sites across the UK. Following further restructuring of the UK civil nuclear industry in 2007, seven site licence companies (owned by separate parent body organisations) were established to work in partnership with the NDA to carry out decommissioning and commercial operations (Fig. 16.2).



16.2 Stewardship of NDA sites.

Other significant producers of radioactive waste in the UK, as owner operators of nuclear licensed facilities, are currently EDF Energy, the Ministry of Defence, GE Healthcare Ltd and Urenco UK Ltd. EDF Energy currently operates advanced gas-cooled reactor (AGR) power stations at seven sites across the UK in addition to one pressurised water reactor (PWR) at Sizewell B. The NDA has an additional responsibility to scrutinise EDF Energy's site decommissioning plans (NDA, 2011).

The UK strategy has been developed to present an integrated approach to the management of RAW and the decommissioning process. All nuclear installations in the UK have been regulated through the Health and Safety Executive (HSE) Nuclear Installations Inspectorate using a site licensing system that applies conditions to operations carried out at the site. The Office for Nuclear Regulation (ONR) is the new regulator for the civil nuclear industry in the United Kingdom. Created on 1 April 2011, the ONR was formed from the merger of the Health and Safety Executive's Nuclear Directorate (the Nuclear Installations Inspectorate, Office for Civil Nuclear Security, and the UK Safeguards Office) and the Department for Transport's Radioactive Materials Transport Team. The change follows the recommendations of a review conducted on behalf of the Government in 2008 (Nuclear Regulatory Review, 2008; HSE and ONR, 2011). The ONR was initially created as a non-statutory body and an agency of the HSE; however, the government has announced its intention to put the ONR on a statutory basis once the appropriate legislation has been passed. When fully operational as a statutory corporation, ONR will be an autonomous organisation, legally separated from, but still supported by, the HSE.

The disposal of RAW has been legislated under the Radioactive Substances Act 1960 (RSA60) and subsequently the Radioactive Substances Act 1993 (RSA93) before being incorporated into Schedule 23 of the Environmental Permitting (England and Wales) Regulations 2010 (EPR10) within England and Wales (RSA93 was retained in Scotland and Northern Ireland). EPR10 was amended in 2011 by the Environmental Permitting (England and Wales) (Amendment) Regulations (2011) to include revised exemption provisions and corresponding amendments were made to RSA93 in Scotland and Northern Ireland. The Environment Agency is the regulator with the responsibility for regulating the disposal of RAW in England and Wales. Responsibility for regulating RAW disposal in Scotland lies with the Scottish Environment Protection Agency, and with the Northern Ireland Environment Agency in Northern Ireland.

16.3 Sources, types and classification of wastes

16.3.1 Waste types

Radioactive wastes may arise as solid, gaseous and liquid materials with different chemical forms. Solid waste streams include organics, metals, concrete and other building wastes, graphite, etc. Liquid waste streams are generally segregated into aqueous wastes and contaminated oily wastes and solvents.

Different waste properties influence the management and disposal options that may be pursued. In general, the UK follows a principle of 'concentrate and contain' when managing wastes. This tends to promote management options for conversion of liquid or gaseous wastes to solid wastes (e.g., through filtration, evaporation, precipitation, encapsulation, vitrification, etc.). The prioritisation of waste management options is discussed further in Section 16.5.1.

16.3.2 Waste classification

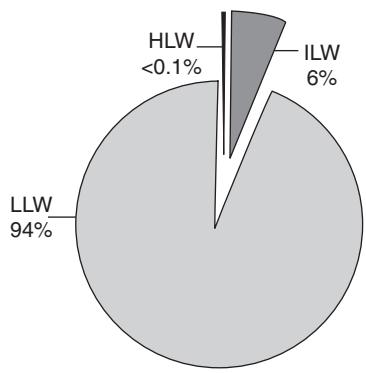
Radioactive wastes in the UK are categorised into low level waste (LLW), intermediate level waste (ILW) and high level waste (HLW) (Defra *et al.*, 2007). In addition, some materials and wastes are defined as out of scope or exempt from the requirements of EPR10 (as amended) in England and Wales (Defra *et al.*, 2011) even though they contain some radioisotopes. Effectively, 'out of scope' equates to 'not radioactive' for the purposes of the legislation. Radioactive substances that are 'out of scope' are not subject to any regulatory requirement under this legislation. Other substances, which are considered to be radioactive by definition, may be exempt from the need for a permit if the level of radioactivity is below the level specified in the

exemption order; however, specified conditions must be met. The levels of radioactivity that are defined as 'out of scope' are taken from EC guidance and are expressed as radionuclide specific activity concentrations.

For naturally occurring radioactive substances or articles used in 'industrial activities', the numerical values are based on a radiation dose of 300 μSv/year to a member of the public. For artificial radionuclides, and for naturally occurring radioactive substances or articles used for their radioactive, fissile or fertile properties (a 'practice'), the values are based on a radiation dose of 10 μSv/year to a member of the public (IAEA, 1988). In effect, this recognises that naturally occurring radioactive materials (NORM) are universal and that it is not practicable to regulate such that the radiation dose criterion of 10 μSv/year to a member of the public is met (IAEA, 2004). Other media and radionuclide specific activity concentrations or total site activity holdings have been established for the exemption order, using the same radiological criteria.

In the case of exemption for disposal, the radiological impact assessments do not assume uncontrolled disposal of waste to the environment. The exemption levels therefore apply to specific types of substance or article (e.g., a waste sealed source), to the disposal route (e.g., to a sewer, or to a landfill), or to the management of waste (e.g., disposed of with considerable quantities of non-radioactive waste), etc. Out of scope and exempt wastes are not considered further here.

Of the radioactive wastes for which a permit is required, LLW is volumetrically the largest component of the UK's radioactive inventory and is classified as waste not exceeding four GBq per tonne of alpha or 12 GBq per tonne of beta/gamma activity (Defra *et al.*, 2007). (The definition of LLW was originally set out in the Government White Paper, Command 2919 (1995) but was superseded by Defra *et al.*, 2007.) Figure 16.3 provides



16.3 Relative volumes of LLW, ILW and HLW.

a breakdown of current and projected RAW arisings in the UK over the next century or so, by waste category. The UK now recognises high and low volume very low level waste (VLLW) as sub-categories of LLW (Defra *et al*., 2007). This offers more flexible, sustainable approaches to long-term management of wastes as alternatives to disposal to the LLW repository (LLWR) at Drigg, Cumbria.

Low volume VLLW is defined by Defra *et al*. (2007) as radioactive waste containing no more than 400 kBq of beta/gamma activity for each 0.1 m^3 and is mostly comprised of small volumes from hospitals and universities. For carbon-14 and tritium-containing wastes, the activity limit is 4,000 kBq for each 0.1 m^3 in total. High volume VLLW is defined by Defra *et al*. (2007) as radioactive waste with an upper limit of 4 MBq per tonne (not including tritium) that can be disposed to specified landfill sites. For tritium containing wastes, the upper limit is 40 MBq per tonne.

ILW is classified on the basis of radioactivity exceeding the upper boundaries for LLW and which does not require heating to be taken into account during storage or disposal. ILW may be sub-categorised as shorter-lived ILW or less radiotoxic ILW. These are not formally defined terms but have been used in a regulatory context to identify wastes that may be suitable for specific waste management options (e.g., Environment Agency *et al*., 2009; Environment Agency and Northern Ireland Environment Agency, 2009).

HLW is waste in which the temperature may rise as a result of radioactive decay and HLW may be referred to as 'heat-generating radioactive waste' (e.g., Defra *et al*., 2008; Command 2919, 1995), although this does not distinguish between types of HLW. HLW in the UK typically arises as a liquid by-product of spent fuel reprocessing. Historical stocks of liquid HLW, together with current arisings, are being conditioned through the Sellafield waste vitrification plant to form a solid material, making it passively safe and suitable for disposal. It is anticipated that by 2015, the UK's HLW will have been converted to vitrified product and will be stored for 50 years to allow further time for radioactive decay.

The term higher activity waste (HAW) has no formal definition and should not be confused with HLW. In Scotland, HAW is used to describe wastes which would otherwise be classified as ILW but which do not generate enough heat for this to need to be taken into account in the design of treatment, storage or disposal facilities (e.g., Scottish Government, 2011) but may also be taken to include LLW which, for one reason or another, is considered unsuitable for disposal as LLW (e.g., Defra and NDA, 2008). The definition of HAW in Scotland is a reflection of the fact that Scotland does not currently possess HLW. The definition of HAW in England and Wales is generally considered to encompass both ILW and HLW in addition to some LLW not suitable for disposal in the LLW repository. Other

radioactive materials may be considered for disposal but are not currently classified as waste. These include spent nuclear fuel and the plutonium and uranium obtained from reprocessing spent fuel (Defra *et al*., 2008).

16.3.3 Sources of waste

Radioactive wastes in the UK can be subdivided according to their sector of origin, notably fuel cycle services, reactor operation, decommissioning activities, nuclear research and non-nuclear licensed site operations such as academic research, radiopharmaceuticals and the medical sector. This breakdown coincides with the approach adopted in the UK discharges strategy (DECC *et al*., 2009), which also recognises wastes from the defence sector and wastes containing NORM. Some wastes (such as HLW) are identified specifically with one or other sector, whilst other wastes (such as LLW) arise at all sites.

Volumetrically, over 90% of the radioactive waste in the UK arises from England. The total amount of waste requiring disposal in the UK, including wastes forecast to arise over the next 100 years, amounts to 4.7 million m^3 or five million tonnes (NDA and DECC, 2011; see also Fig. 16.3). The vast majority of this consists of future waste to be decommissioned from existing facilities.

Of waste produced from existing facilities, nearly 75% originates from the reprocessing of spent fuel. Approximately 15% arises from nuclear power stations and approximately equal amounts in the region of 5% of wastes are related to the research and development sector (a legacy of government-funded programmes dating from the start of the UK nuclear industry) and uranium enrichment and fuel fabrication (NDA and DECC, 2011). Defence-related and medical and industrial wastes account for approximately 1% of that produced from existing facilities. Waste generated from spent fuel reprocessing, carried out at Sellafield, also encompasses waste from legacy waste programmes in the defence sector (NDA Inventory Summary report, 2011).

Over 90% by volume of the UK's wastes are classified as LLW, with 6% comprising ILW and 0.1% HLW (Figure 16.3). This small proportion of HLW contains approximately 95% of the total radioactivity, while LLW, by contrast, is responsible for 0.01% of the overall radioactivity.

16.4 Development of radioactive waste (RAW) management strategies

Regulation and industry practice have evolved to reflect the principles of radiological protection and practicability of implementing options. This section is primarily aimed at the civil nuclear sector. Defence sites and

wastes are generally managed in a similar way, although formal requirements identified for the civil nuclear industry do not extend to defence activities. However, it is the policy of the Ministry of Defence (MoD) to meet standards equivalent to them where practicable.

16.4.1 Application of the best available techniques

The optimisation of processes and implementation measures to reduce industrial discharges has a long history in the UK. The use of best practicable means (BPM) to abate smoke and other stack discharges can be traced back to the Alkali Act (Amendment) 1874, which required that '*the owner of every alkali work shall use the best practicable means of preventing the discharge into the atmosphere of all other noxious gases arising from such work, or of rendering such gases harmless when discharged.*' Use of BPM became a regulatory requirement in various fields and was eventually integrated within the permitting process for managing radioactive wastes.

More recently, the Royal Commission formulated the concept of the best practicable environmental option (BPEO) to minimise total environmental impact in the context of multi-media discharges (RCEP, 1976). The Royal Commission on Environmental Pollution (RCEP)'s Twelfth Report (1988), elaborated on the concept, and defined it as: '*the outcome of a systematic and consultative decision-making procedure which emphasises the protection and conservation of the environment across land, air and water. The BPEO procedure establishes, for a given set of objectives, the option that provides the most benefits or the least damage to the environment as a whole, at acceptable cost, in the long term as well as the short term.*'

Whilst the concept of optimisation has been adopted globally, the BPM/BPEO terminology has not been used outside the UK; and within the UK has not been widely used outside the nuclear sector for some years. The recent introduction of the Environmental Permitting Regulations (EPR) in England and Wales formed part of a major initiative to simplify and reduce the costs of permitting activities. In parallel, there has been a shift in regulation of the nuclear sector to adopt a more uniform approach consistent with other industry sectors. As a consequence, in England and Wales the use of BPM terminology has been discontinued and replaced with use of best available techniques (BAT), although in Scotland and Northern Ireland the use of BPM as an authorising tool will continue in the context of RAW management.

Early discharge authorisations (especially for gaseous emissions) were based on use of BPM with an implied 'dilute and disperse' philosophy and with less emphasis on numerical discharge limits. From the late 1970s numerical limits were increasingly established throughout the industry for

both liquid and gaseous wastes. Over a similar timeframe the identification as BPM, and use of, interim 'delay and decay' storage tanks reduced, as the philosophy shifted to one of 'concentrate and contain' for disposal as solid waste.

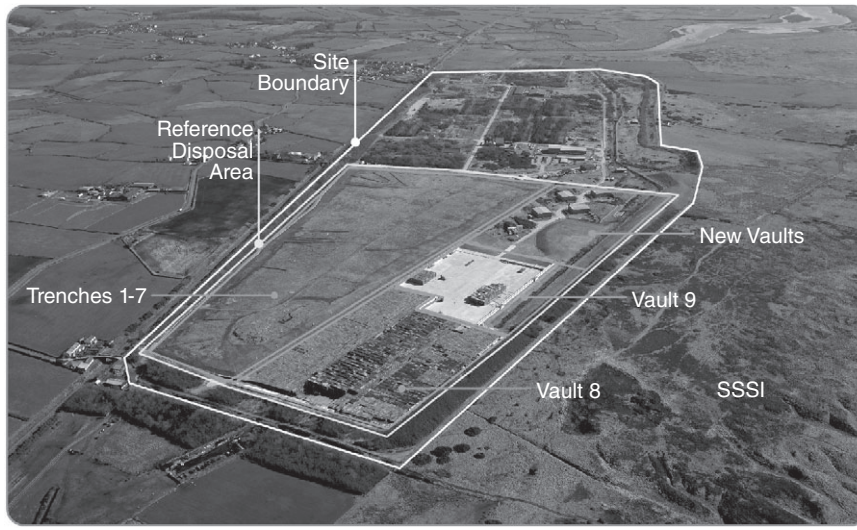
In implementing BAT approaches to RAW management, a number of other principles are taken into account. The proximity principle requires the disposal of solid waste to be as close to its source as possible to minimise the environmental consequences of transportation. Proportionality is also a central theme in the application of BAT: the cost of implementation must be in proportion to the benefit of its introduction. In fact, the Environment Agency guidance states that all reasonable steps must be taken to reduce the doses to people unless the costs are 'grossly disproportionate' to the benefits (Environment Agency, 2010).

16.4.2 Integrated waste management strategy

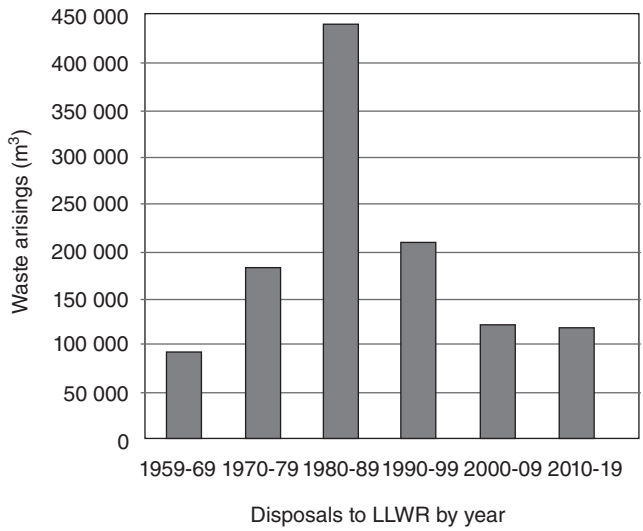
UK government policy requires that there is effective management of all RAW in the UK. An integrated waste strategy (IWS) is one of the requirements of site licence operators working under the auspices of the NDA, and the environment agencies and HSE are encouraging other nuclear sites to do so as well (SD:SPUR, 2007). (If an IWS is not prepared, a site waste management plan is required for new construction sites with costs in excess of £300,000.) Development and maintenance of an IWS is one of the standard requirements attached to radioactive substances activity permits issued by the Environment Agency under EPR10 (as amended). The NDA's overall strategy for RAW management of the bulk of the UK's civil nuclear sites is reviewed every five years in response to a requirement of the Energy Act (2004).

For LLW, the primary strategic aim is to reduce the amount of waste being disposed of to reduce overall NDA liabilities and costs by retaining future capacity of the UK's primary LLW repository, the LLWR, at Drigg, Cumbria (Figs 16.4 and 16.5).

Strategy development for the UK's higher activity wastes has evolved following recommendations from the independent Committee on Radioactive Waste Management (CoRWM) to the UK government in 2006 (CoRWM, 2006). Government in England and Wales accepted the recommendations for developing safe and secure methods for interim storage of HAW, coupled with an ongoing research and development programme prior to the development of a geological disposal solution for the permanent disposal of the wastes. The devolved administrations' position on HAW is slightly different. The Welsh Assembly Government chose to reserve its position on geological disposal whilst acknowledging the CoRWM recommendations. Scotland's HAW policy, set out in a recent document



16.4 The LLWR site in March 2010. SSSI is a site of special scientific interest due usually to its rare flora and fauna. Reproduced with permission of LLWR Ltd.



16.5 Historical and projected disposals to LLWR by volume, 1959–2019.

(Scottish Government, 2011), outlined the preferred option for the long-term management of HAW in near-surface facilities near to the source of the waste.

UK government agreed with the CoRWM recommendations for the geological disposal facility to be permanently sealed once operations ceased, although the policy recognises that the final decision on this remains flexible. The NDA's revised strategy is structured according to a number of themes to focus future development: site restoration, spent fuels, nuclear materials, integrated waste management, business optimisation and critical enablers. Of these, site restoration, supported by integrated waste management, are considered to be the driving forces.

16.4.3 Guidance for permitting requirements for waste disposal

Applications for approval of RAW disposal facilities made to the environment agencies and planning authorities under EPR10 (as amended) must be supported by an environmental safety case. Requirements for authorisation are set out in a guidance document published jointly by the environment agencies, 'Near-surface Disposal Facilities on Land for Solid Radioactive Wastes: Guidance on the Requirements for Authorisation' (the GRA). This was originally published in 1997 (Environment Agency *et al.*, 1997) and revised and updated in 2009 (Environment Agency *et al.*, 2009). An equivalent document sets out the guidance for disposal of geological waste, although the guidance for deep geological disposal does not apply in Scotland. (Environment Agency and Northern Ireland Environment Agency, 2009).

The Environment Agency has also initiated a consultation, and published draft guidance, on the setting of limits for disposal of liquid and gaseous RAW into the environment under EPR10 (as amended) (Environment Agency, 2011).

16.4.4 Sea disposal

Between 1949 and 1982 about 33,000 m^3 of RAW was disposed of in the North Atlantic and UK coastal waters (NDA and DECC, 2011). The London Convention 1972 prohibited all major nuclear powers from disposing HLW at sea although low level and intermediate level waste disposal continued into the 1980s* when a voluntary moratorium came into force. Sea disposal of solid radioactive waste was abandoned by the UK in 1983.

*The London Convention subdivided radioactive waste into high and low level waste, with definitions of high and low level waste that were derived specifically for disposals at sea.

The Rio Declaration on Environment and Development (1992) reaffirmed the voluntary moratorium on sea disposal and the London Convention of 1996 brought into effect the precautionary approach with a ban on all LLW disposals at sea.

16.4.5 On-site disposal

The Sellafield site has had its own on-site licensed landfills for disposal of waste throughout the operation of the plant, initially in trenches within the Separation Area in the 1950s (before development of the LLWR) and then in mounds built up on the coastal fringe and on the northern perimeter of the site. Two sites remain in operation, the south landfill site and the Calder Plain landfill extension. These primarily receive low level radioactive soil (BNFL, 1976–2004).

On-site waste disposal has been undertaken at other sites in the UK (notably Hunterston and Dounreay in Scotland), but this practice has not been widespread within UK nuclear licensed power generation sites.

16.4.6 UK strategy for discharges

In 2009, UK government published a revised UK Discharge Strategy, which updates government policy and describes how the UK will continue to implement the agreements reached at the 1998 OSPAR Convention, and subsequent OSPAR meetings on radioactive substances, particularly the radioactive substances strategy. This builds on the initial UK Strategy, published in 2002, and expands its scope to include aerial, as well as liquid discharges, from decommissioning as well as operational activities, and from the non-nuclear as well as the nuclear industry sectors. The objectives of the strategy are:

- to implement the UK's obligations, rigorously and transparently, in respect of the OSPAR Radioactive Substances Strategy (RSS) intermediate objective for 2020; and
- to provide a clear statement of government policy and a strategic framework for discharge reductions, sector by sector, to inform decision making by industry and regulators.

The expected outcomes by 2020 are:

- progressive and substantial reductions in radioactive discharges (to the extent described in the strategy);
- progressive reductions in concentrations of radionuclides in the marine environment resulting from radioactive discharges, such that by 2020 they add close to zero to historical levels; and

- progressive reductions in human exposures to ionising radiation resulting from radioactive discharges, as a result of planned reductions in discharges.

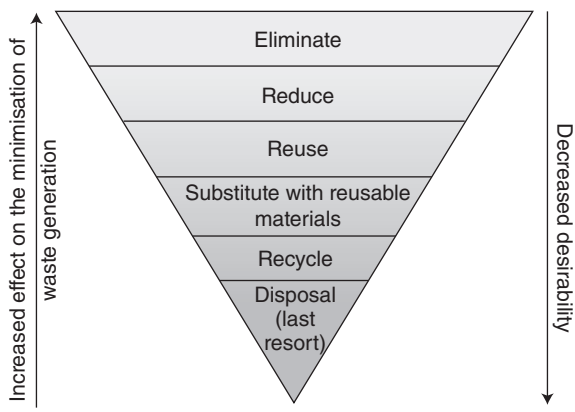
16.5 Current RAW management practices and strategies

16.5.1 Waste hierarchy and waste minimisation

A waste hierarchy is essentially a proactive management policy that prioritises waste management options, with the overall aim of reducing the amount of waste that is ultimately disposed. The principle of the waste hierarchy has been the foundation of waste management policy for decades following its first appearance in EU policy in the mid-1970s and plays a central role in EU waste policy (Article 4 of the Waste Framework Directive). However, its introduction to RAW management did not take place until 2006–2008. The waste hierarchy (see Section 1.5.2) is summarised in Fig. 16.6.

Progression of the waste hierarchy towards the minimisation of the amount of wastes disposed does not necessarily go hand in hand with the most sustainable environmental option. Waste hierarchies are often implemented in a complementary way with BAT to ensure that sustainable approaches are integrated into the overall strategy. It is also implemented in combination with a life cycle approach, which affects every stage from design, construction, operation and decommissioning of disposal facilities.

Because of the considerable volume and weight reduction involved in combustion, low level wastes in the UK, particularly plastic, cellulose products and oil wastes, are often incinerated. Incinerators accepting LLW and high volume VLLW are licensed under EPR (2010) for disposal of



16.6 The waste hierarchy as applied in England and Wales.

radioactive waste. Low volume VLLW is exempt under EPR (2010) as radioactive waste, but the operator of an incinerator will still require permission for non-radioactive waste incineration.

16.5.2 Low level waste

LLW disposal in the UK has been ongoing since the 1950s, providing considerable perspective on the approaches to, and the strategy development of, LLW disposal. Since 1959, LLW has been disposed of at the UK's national low level waste repository (LLWR) in Cumbria, in addition to a number of other LLW disposal sites including various Sellafield pits, Hunterston A and Dounreay. LLW strategy development has largely progressed in response to the need to preserve the capacity of the LLWR for as long as possible given future arisings of LLW in the UK.

The LLWR (Fig. 16.4), located on the site of a second World War munitions factory, was initially developed as a series of excavated trenches into which wastes were loose tipped between 1959 and 1995 (LLWR, 2011). The trenches were designed with drainage and runoff collection systems and were largely keyed into a low hydraulic conductivity clay layer. Where this was absent, bentonite was rotovated into the trench base.

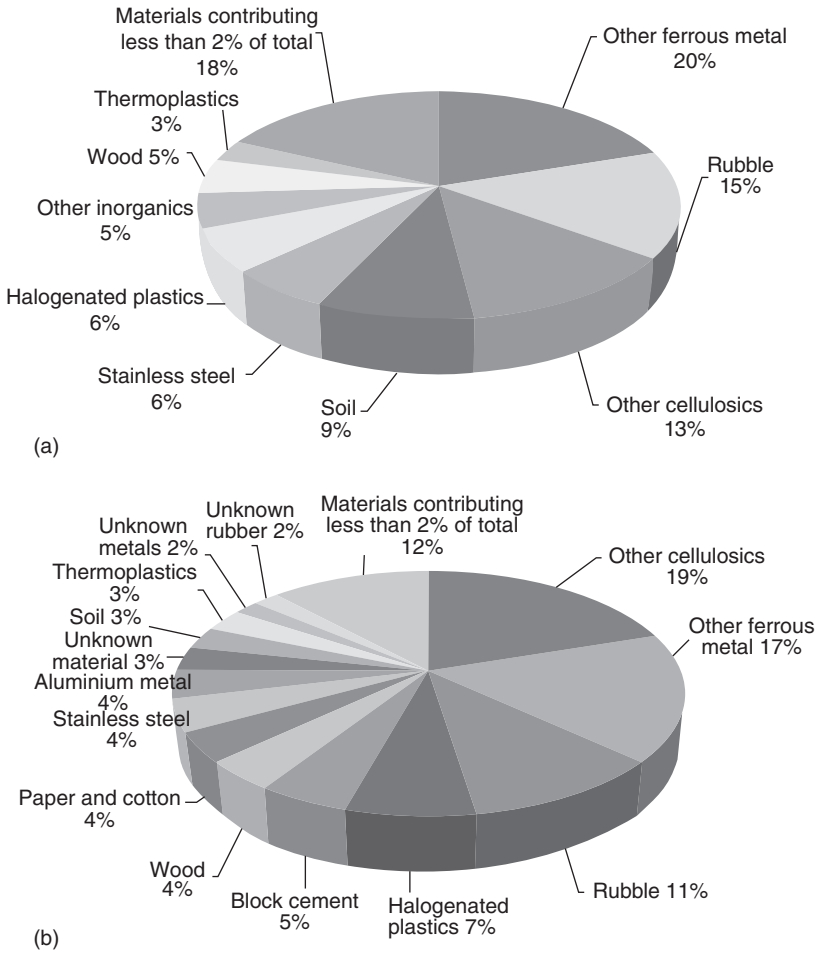
In 1988, trench disposals were phased out in favour of LLW disposal to engineered vaults. A number of improvements were made to the trench disposal areas after this time. The installation of an interim cap over Trenches 1–6 took place in 1989 to minimise rainfall ingress into the wastes and a bentonite cut-off wall was excavated on the north and east sides of the disposal area in 1988 to reduce the potential for tritium migration in groundwater (LLWR, 2011).

Overall, the major components of LLW are building rubble, soil and steel items from the dismantling and demolition of nuclear reactors and other nuclear facilities (Fig. 16.7).

Very low level waste

A policy statement for the long-term management of solid low level radioactive waste in the United Kingdom was published by Defra and the devolved administrations in 2007 (Defra *et al.*, 2007). Among other provisions, this identified a need for greater flexibility in the approach to LLW disposal and, subject to permitting based on a 'risk informed approach', applications may be made to dispose of LLW/VLLW to conventional landfill sites (inert, non-hazardous and hazardous waste disposal sites).

The first site to have been licensed for high volume-VLLW disposal to landfill is the Lilyhall landfill site operated by Waste Recycling Group and Energy*Solutions*. Some of these proposals have been controversial with



16.7 Main materials in the LLWR (a) trenches and (b) vaults to March 2008.

local communities. Following an appeal against a rejection of planning permission by Northamptonshire County Council, permission for LLW disposal was granted for another site at King's Cliffe, operated by Augean.

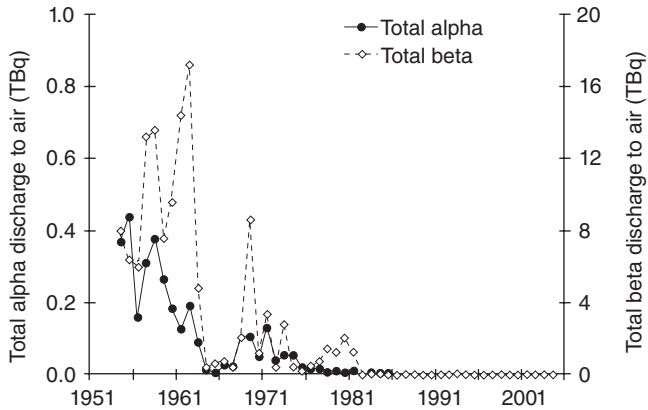
Introduction of waste treatment for LLW

Where wastes cannot be prevented from arising, the UK strategy is aimed at minimising the volume and activity of LLW consigned for disposal through waste segregation, decontamination or decay storage. A variety of strategies are encompassed in recycling; including waste volume reduction and compaction and thermal treatment. Reuse and recycling of rubble and

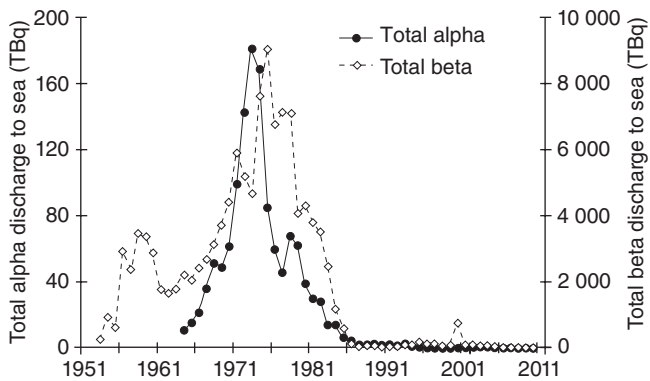
metal wastes provide a further means to reduce LLW wastes. Metallic drum wastes from the LLWR are currently being shipped to the Studsvik facility at Lillyhall, Cumbria for processing to remove the radioactivity to minimise the quantities of metals disposed in the LLWR. Suitable LLW is also super-compacted under high pressure before disposal at the LLWR to minimise the volume prior to disposal.

Discharges

Discharges from Sellafield (see Figs 16.8 and 16.9, compiled from Gray *et al.*, 1995; Jackson *et al.*, 2000; Environment Agency *et al.*, 1971–2011 and



16.8 Total alpha and total beta discharges to air from Sellafield, 1951–2010. Note that total alpha and total beta are control measures with defined meanings under the terms of site permits.



16.9 Total alpha and total beta discharges to sea from Sellafield, 1951–2010. Note that total alpha and total beta are control measures with defined meanings under the terms of site permits.

BNFL, 1976–2004) illustrate the influence of many factors affecting LLW management, including perception of tolerable risk, the design and specification of new plant and post-operational waste conditioning, and the changing emphasis to reduce discharges to the environment in favour of solid waste disposals.

Peak discharges to air occurred in the 1960s and peak discharges to sea occurred in the 1970s. Over the past decades there has been an increasing emphasis on effluent treatment and containment of radioactivity within solid wastes. At the same time, there has also been an increasing emphasis on reducing waste arisings and on volume reduction for those wastes that cannot be avoided. The rise in discharges to sea during 2001 reflected primarily processing of larger quantities of medium active concentrates (with associated increased discharges of Tc-99 and Sr-90). Discharges of C-14 to sea also increased in 2001, mainly due to diversion (by introduction of a gas scrubber) of activity previously discharged to air, recognising that this diversion was made to reduce the overall environmental impact of site discharges (BNFL, 2002).

16.5.3 Intermediate level waste

Waste management strategies for ILW are currently variations on interim storage of unconditioned or conditioned (e.g., grout encapsulated, polymer encapsulated or vitrified) waste. For higher activity waste, government policies for England and Wales differ from those of Scotland. ILW strategy in England and Wales is aimed at providing storage facilities prior to development of a geological disposal facility (GDF). The objective is for safe storage to be possible for 100 years or more and for the strategy to be revised if a geological disposal facility is not available by 2100. In Scotland, the policy for the long-term management of HAW is aimed at storage in near-surface facilities that are located as near as possible to the site where the waste was produced (Scottish Government, 2011). Recognising the uncertainty in future developments of the strategy for HAW, long-term monitoring and the capability of retrieval of the wastes is a requirement of the Scottish approach. Within this framework, the NDA is committed to reducing risks by dealing with higher activity wastes stored in ageing, legacy facilities and placing them into safer storage conditions.

There are currently a number of ways that ILW is stored in the UK:

- raw, untreated waste in legacy facilities;
- historically treated waste in storage that would require further treatment or conditioning before long-term safe storage could take place;
- interim storage of conditioned waste (in cement);

- waste stored in modern facilities that would still require waste conditioning;
- in-situ waste awaiting decommissioning.

ILW is being treated and conditioned ready for disposal, despite the unavailability of a GDF. Such treatment and conditioning is in accord with generic repository assessments undertaken by the radioactive waste management directorate (RWMD) of the NDA. The RWMD operate a Letter of Compliance process to advise waste producers concerning the suitability of their packaging and conditioning approaches. In general, the UK's ILW is encapsulated in cement and contained within stainless steel or concrete containers (Ojovan and Lee, 2005). The Letter of Compliance process is regulated by the ONR and the environment agencies under joint regulatory arrangements.

In 2009, there were 19 modern ILW stores in the UK. The majority of the largest hazards and environmental risks posed by ILW in the UK are stored at Sellafield and Dounreay. The NDA's strategy is focused on reducing these risks by removing wastes to newer and safer facilities (NDA, 2009). The waste is conditioned prior to storage and, as of 2008, 8% of the UK's ILW had been conditioned, packaged and placed into interim storage (NDA, 2009).

The bulk of the UK's ILW, mainly Magnox fuel cladding held in underwater tanks at Sellafield, was until recently stored in a series of 22 silos. The contaminated liquid has presented a challenge to decommissioning and retrieval of the waste. In 2010, the liquid was piped in a shielded structure over the Sellafield site to the Sellafield ion exchange effluent plant (SIXEP) processing facility, where the radioactivity was removed by filtration over aluminosilicate sand. The intention is to store this ILW in solid form, while the silos are replenished with clean water and repeatedly diluted.

16.5.4 High level waste

The UK's HLW arising from reprocessing of spent fuel using the Plutonium URanium EXtraction (PUREX) process is immobilised by vitrification. In addition, it is expected that some spent fuel from the UK's advanced gas-cooled reactors (AGRs) will be directly disposed of along with some Pu immobilised in ceramics as well as spent fuel from any new generation of reactors.

No facilities for disposing of ILW and HLW have been developed; these wastes are currently stored. LLW not suitable for near-surface disposal is also stored. It is anticipated that this waste will be disposed to a facility developed for ILW and HLW disposal.

In 2001 the Managing Radioactive Waste Safely (MRWS) programme was initiated by the UK government and the devolved administrations to take a fresh look at the management policy of the UK's higher activity

wastes. The UK government published a White Paper in June 2008 (Defra *et al*., 2008) setting out its detailed policy and plans for the long-term management of higher activity wastes.

The White Paper sets out an approach based on voluntarism and partnership with local communities, coupled with the use of appropriate site screening and assessment criteria as the basis for siting a GDF (repository). Overseas experience (e.g. Chapter 13) suggests that such an approach is likely to be an effective way of addressing the concerns of communities about hosting such a facility.

The UK government invited communities to express an interest in taking part in discussions about the siting process for a geological disposal facility, and from 2010–13 two borough councils (Copeland and Allerdale in Cumbria) as well as Cumbria County Council (CCC) expressed an interest in hosting a GDF forming a partnership for discussions with government and the local communities. However, in January 2013 CCC decided against participating further in the process and the UK government is currently consulting on ways of encouraging further communities to come forward. Historically, most of the HLW in England and Wales has arisen at Sellafield, which is sited in the borough of Copeland, Cumbria. The NDA is the implementing organisation, responsible for planning and delivering the GDF and, as part of this process, will work with government to engage with communities and other stakeholders.

The NDA and its agents will have the responsibility for securing the necessary regulatory and planning permissions involving any host community and planning authorities as necessary.

16.5.5 Other materials

The MRWS programme also takes into consideration some radioactive materials that are not classified as wastes in the UK. These materials include uranium, plutonium and some spent nuclear fuel associated with civil nuclear activities. They have potential value: uranium and plutonium can be used to make nuclear fuel, and spent nuclear fuel can be reprocessed to recover uranium and plutonium for reuse. However, if it was decided at some point in the future, on the basis of economics or environmental and safety issues, that these materials had no further use, they may need to be managed as wastes. Radioactive materials that are not deemed to be waste are not reported in the UK Radioactive Waste Inventory, but summary information is provided in a separate document that is published with the Inventory.

16.5.6 Implementation of geological disposal

The UK government (through the Department of Energy and Climate Change, DECC) is working with the NDA through its RWMD (a prospective

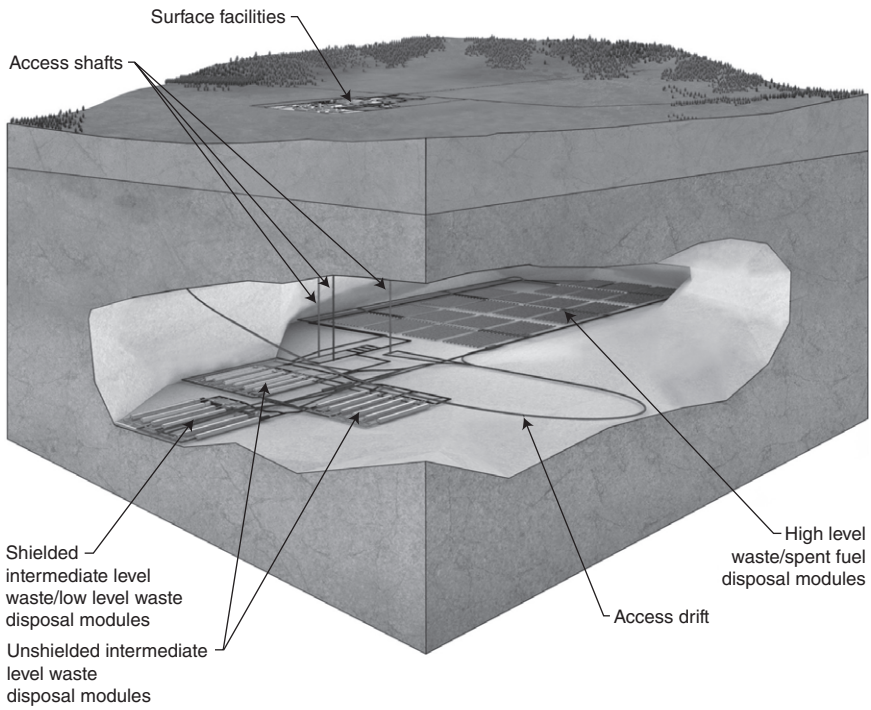
site licence company) on finding a site and implementing the construction of a GDF through the volunteer process. The variety of the UK's wastes mean that a multi-level and chamber repository will be needed with different wastes in separate sections (Fig. 16.10; NDA, 2010).

16.6 Contaminated site clean-up

16.6.1 Regulation

Decommissioning represents a current challenge to the UK nuclear industry because many of the UK's reactors have reached the end of their useful life.

Nuclear site licence holders are required to prepare strategies for decommissioning of nuclear infrastructure. The Nuclear Reactors (Environmental Impact Assessment for Decommissioning) Regulations 1999 (EIADR99) as amended by the Nuclear Reactors (Environmental Impact Assessment



16.10 Schematic illustration of the layout of a generic GDF (not host-rock specific) for all UK higher activity radioactive wastes. Figure courtesy of Nuclear Decommissioning Authority.

for Decommissioning) (Amendment) Regulations 2006 (EIADR06) require that nuclear reactor decommissioning is accompanied by an Environmental Impact Assessment (EIA) in addition to other requirements. The purpose of EIADR is to require assessment of the potential environmental impacts of projects to decommission nuclear power stations and nuclear reactors. In addition, the intention is to make the decision-making process open and transparent. EIADR require that the public and other relevant stakeholders be consulted from an early stage, regarding the environmental impacts of the options being considered for a proposed decommissioning project.

An Environmental Statement containing details of potential environmental impacts must be submitted to the Health and Safety Executive before any decommissioning work can be carried out, and once they have consulted with various bodies, i.e. the environment agencies, conservation bodies, local authorities and stakeholders, permission may be granted.

HSE have developed policy (HSE, 2005) and guidance (HSE, 2008) on the criteria to be satisfied for sites to be de-licensed. The basis is the demonstration of 'no danger' remaining from the presence of any remaining contamination on the site. The requirement is 'to show that any such remaining radiological hazard will not pose a significant residual risk to any person, for all reasonably foreseeable uses to which the site may be put and not just for its next future use. Based on the reasoning laid out in the HSE publication "Reducing Risk and Protecting People", HSE believes that the annual risk of a fatality of one in a million to an individual is regarded by society as "broadly acceptable".'

HSE (2008) also refers to the need to show that risks are ALARP (as low as reasonably practicable). HSE's preferred method for demonstrating that the risk criterion has been met is the application of derived concentration levels for the clearance and exemption of radioactive substances (IAEA, 2004). These values are calculated on the basis of a dose criterion of 10 μSv/y and a set of exposure scenarios.

The management of radioactively contaminated land on a nuclear licensed site is carried out by ONR. ONR regards radioactively contaminated land and emplaced radioactive substances on nuclear licensed sites as accumulations of nuclear matter, unless they are, or arise from, authorised disposals, and it requires licensees to manage it as such. The licence conditions require that licensees control or contain nuclear matter, to record the amount of radioactive material and its location, and justify and demonstrate the arrangements to maintain safety by means of a safety case.

For radioactively contaminated land that is not on a nuclear licensed site, a different set of regulations apply. Part IIA of the Environmental Protection Act 1990 provides a regulatory regime for the identification and remediation of contaminated land that is causing unacceptable risks to human health or the wider environment. In 2006 and 2007 this was extended to

cover radioactivity and to cover land contaminated with radioactivity originating from nuclear installations (different regulations were enacted in England and Wales and in Scotland). The objectives for the extension of Part IIA to radioactive contamination remain the same: to provide a system for the identification and remediation of land where contamination is causing lasting exposure to radiation for human beings and where 'intervention' is liable to be 'justified'. A key aspect is that the land should be 'suitable for use'. The criteria for designating land as 'radioactively contaminated land' are based on a probability weighted annual dose of 3 mSv from the contamination. In the case of contamination in the form of discrete radioactive particles that could give rise to doses above 50 mSv if encountered, decisions on whether the land should be designated as radioactively contaminated land are based on a number of factors.

In the context of new development of land, radioactive contamination may also be deemed a material planning consideration under the relevant Town and Country Planning Act.

16.6.2 Successful clean-up of radiological contamination

The Harwell Nuclear Licensed Site in Oxfordshire has successfully cleaned up and delicensed part of the site. The site was a former RAF airfield before it was used for research associated with the development of nuclear power in the UK. The original licensed site was 113 hectares, containing four research reactors. A phased approach to delicensing was adopted based on the programme for decommissioning the facilities. In 1992 five hectares were delicensed and in 2006 a further seven hectares, originally containing 43 buildings, were delicensed. Ten of the buildings had been used for work involving radioactivity. The facilities were decommissioned and the land and buildings certified free of ionising radiation and available for non-nuclear development. A further five hectares, including the former site of the research reactor GLEEP (the graphite low energy experimental pile), were delicensed in 2011. In this instance, all the buildings were demolished and some concrete foundations were left. The case for delicensing a further five hectares has been submitted. Experience gained in the delicensing work was that it is important to pay attention to detail and to work with the ONR as far as possible. It is best practice to build delicensing requirements into decommissioning and land remediation works and to keep good decommissioning and remediation records. Often it is important to demonstrate the absence of something, for example that the section of drain is not there any more.

Since 1984, a programme of monitoring for radioactive objects has been carried out on beaches in the vicinity of the Sellafield site in West Cumbria. During this programme, over 650 radioactive objects were identified and

removed up to the summer of 2009, comprising particles with sizes smaller or similar to grains of sand and also contaminated pebbles and stones. These objects have a much higher activity content that can be easily distinguished from the ambient homogeneous levels of contamination on the beaches. The source of these objects is not known but there have been a number of known events in the past that have resulted in release of radioactive particles into the environment, including early operation of the Windscale piles (1952 to 1957), the Windscale fire in 1957 and the beach incident in 1983. Hence the management strategy for the clean-up of the beaches has to consider a wider context than just the beaches, e.g., the terrestrial and marine environment.

16.6.3 Problematic cases and lessons learned

The Sellafield nuclear licensed site on the West Cumbrian coast has a history of over 60 years of industrial activity, starting with its wartime development as an explosives factory and moving on to cover an extensive range of nuclear processing and power generation operations. The area around the Magnox reprocessing plants and the older production plants in the centre of the site is known as the 'Separation Area'. This area was greatly expanded in the mid-1960s but has not been significantly altered since.

A number of leaks of radioactive liquors to ground are known to have occurred from several plant buildings, waste storage vaults and burial trenches within the site, largely associated with the older process plant in the Separation Area. Records from boreholes and other excavations undertaken during three decades of site engineering and construction work have demonstrated that radioactively contaminated ground exists beneath parts of the Separation Area and occasionally in the wider site. In addition to radioactivity, there is the potential for other components of spent fuel reprocessing to have contaminated ground, including inorganic salts from neutralised acids, solvents and other organic compounds as well as the more typical contaminants normally associated with industrial activity, such as heavy metals, fuel, oils, etc. The site has had its own on-site landfills for disposal of waste throughout the operation of the plant, initially in trenches within the Separation Area in the 1950s and then in mounds built up on the coastal fringe and on the northern perimeter of the site.

In 2002 a major investigation was started at Sellafield, in order to better understand the extent of contaminated land and groundwater at the site (BNFL, 2004). The first phase of this investigation involved the drilling of 73 boreholes on- and off-site, outside of the Magnox fuel reprocessing area. In 2007 a structured programme of work was initiated to develop and calibrate a model of groundwater flow on the Sellafield site, which could be

used to underpin future land quality programmes of work. Since then, a borehole drilling programme comprising a total of 3,124 m with 138 permanent groundwater installations has been completed.

Migration of contaminants occurs in the groundwater. In addition to the natural characteristics of groundwater flow, the possible impact of features such as drains and building foundations as preferential pathways or barriers to flow is likely to be significant. The regional groundwater flow direction is from the high ground inland towards the coast in the south-west, but local variations appear to exist, in particular an apparent westerly groundwater flow along the axis of a buried, sandstone-infilled channel under the Separation Area. The dominant radioactive contaminants in groundwater are strontium-90 (^{90}Sr), caesium-137 (^{137}Cs), tritium (^{3}H) and technetium-99 (^{99}Tc).

An estimate of the total volume of soil contaminated with radioactivity above natural background levels has been made of the order of 13 million m^3. Of this estimated volume of contaminated soil, over 90% has been described at Sellafield as VLLW, one million m^3 is in the LLW category and an estimated 1,600 m^3 is ILW, based on known information of radioactive leaks in the Separation Area.

The potential for contamination of land and groundwater in this area is currently an item of uncertainty with regard to liability cost estimates for the Sellafield site. This work will enable future estimates to be made with greater confidence. It will also support ongoing safety and environmental management and aid the development of strategy and planning for the future.

16.7 Sharing experience

The Environment Agencies' Requirements Working Group (EARWG) was established in 2003 to share information regarding best practice in RAW minimisation. An objective of the UK LLW Management Plan is to identify and share waste minimisation practices in order to minimise the burden on the environment from disposal of radioactive wastes at the LLWR. In addition to minimising waste disposals to the LLWR, the use of recycled materials rather than virgin resources is preferable because it saves energy, reduces emissions of greenhouse gases and other air and water pollutants and, of course, conserves natural resources.

There are a number of factors, including economic, regulatory and availability that make the re-use and recycling of materials previously classified as solid RAW a challenging task. Nonetheless, segregation by decontamination or physical removal may enable radioactive material to be removed from the bulk of low radioactivity material (i.e. high volume low activity or

exempt material). This means that only a relatively small volume of material needs to be classified as RAW, whilst the bulk of the low radioactivity material has the potential to be re-used or recycled. There is a range of physical, chemical, electrochemical and dismantling techniques that result in the segregation of solid material.

Both EARWG and the LLWR Strategy Group maintain websites to share information.

The SAFEGROUNDS (SAFety and Environmental Guidance for the Remediation of contaminated land On UK Nuclear and Defence Sites) learning network was established in 1998 and provides a forum for developing and disseminating good practice guidance on the management of radioactively and chemically contaminated land on nuclear and defence sites in the UK. SAFEGROUNDS is now well established and shares information via its website.

Partly arising from SAFEGROUNDS, the SD:SPUR (Site Decommissioning: Sustainable Practices in the Use of Resources) learning network was established in 2004 to develop through dialogue safe, socially, economically and environmentally sustainable practices in the use of resources arising from the decommissioning of nuclear sites. The project has published guidance on the potential applications for the re-use and recycling of these wastes, and the factors controlling their supply and demand, and has developed a set of sustainability indicators that could be used by site operators when identifying and choosing between options for the management of these wastes. Information from SD:SPUR is shared freely through its website.

16.8 Future trends

16.8.1 International commitments

In 1998 the contracting countries to the OSPAR convention of 1992 adopted the Sintra statement (OSPAR Commission, 1998). The aim of this was to achieve substantial reductions in discharges, emissions and losses of radioactive substances to near background levels for naturally occurring radioactive substances and to near zero for artificial radionuclides. The Sintra statement was released with the objective of reducing radioactive discharges to a level where additional concentrations were close to zero by the year 2020. This presents challenges for both decommissioning of sites (which may result in transient spikes in discharges as a result of post-operational plant clean out and other practices) and to reprocessing or other waste management practices which lead to high volumes of very low level liquid effluents.

16.8.2 Approaches to waste management and disposal

The top priority for the NDA in England and Wales remains the higher hazard facilities at Sellafield, especially those associated with legacy plant and historical high level wastes, and the development of a geological disposal facility.

Alternative disposal options for ILW have been explored and may present safe and economic facilities for use at site or national level. In addition, the current unavoidable extended storage of higher activity wastes may result in the potential for reclassification of some wastes due to radioactive decay or volume dilution arising from unavoidable dilution due to waste conditioning processes. This presents potential challenges to regulation of waste management practices.

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17

Scotland: experience of radioactive waste (RAW) management and contaminated site clean-up

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Abstract: This chapter discusses the nuclear operational, nuclear decommissioning and associated radioactive waste activities being undertaken at the eight nuclear sites in Scotland. The Scottish government radioactive waste policies governing the sites are explained and put into context with those of the rest of the UK. The history, current activities being undertaken, and the anticipated future programmes are described for each site. The scales of the programmes are illustrated by giving summary data of the volumes of radioactive waste involved and the timescales over which it is to be managed. Brief mention is made of radioactive waste associated with the non-nuclear industry.

Key words: Scottish radioactive waste policy, volumes of radioactive waste in Scotland, decommissioning strategy, disposal and storage of radioactive waste, nuclear power in Scotland.

17.1 Introduction

Nuclear installations for both civil and military purposes began to be built in Scotland from around 1950. Some remain operational today, others are undergoing decommissioning, and new facilities are being constructed at some of the sites. Although located in Scotland, no sites are Scottish owned. The military installations are owned by United Kingdom (UK) government, Ministry of Defence (MoD). The operational nuclear power stations are owned by multinational companies. The installations undergoing decommissioning are owned by UK government, Nuclear Decommissioning Authority (NDA).

There are eight separate nuclear sites at six locations in Scotland and these are described in Table 17.1 and their locations shown in Fig. 17.1.

All the nuclear installations create and manage radioactive wastes (RAW), though there is a wide range in the volumes and categories of waste managed at the different locations.

Permission has been given by the NDA for the use in this chapter of NDA and Department of Energy and Climate Change (DECC) copyright source data on waste volumes in the NDA 2010 UK Radioactive Waste Inventory

Table 17.1 Scottish nuclear sites

Owner and site name	Site operating manager	Location	Operations at site
NDA			
Dounreay	Dounreay Site Restoration Ltd	Caithness	Decommissioning and LLW disposal
Hunterston A	Magnox	Ayrshire	Decommissioning
Chapelcross	Magnox	Dumfries and Galloway	Decommissioning
EDF Energy			
Hunterston B	British Energy	Ayrshire	Power station
Torness	British Energy	East Lothian	Power station
MoD			
Naval Reactor Test Establishment (NRTE) Vulcan	Rolls Royce	Caithness	Testing nuclear submarine propulsion reactor
Her Majesty's Naval Base (HMNB) Rosyth Royal Dockyard	Babcock International	Fife	Decommissioning submarine facilities
Her Majesty's Naval Base (HMNB) Clyde	MoD	Dunbartonshire	Support to operational submarines

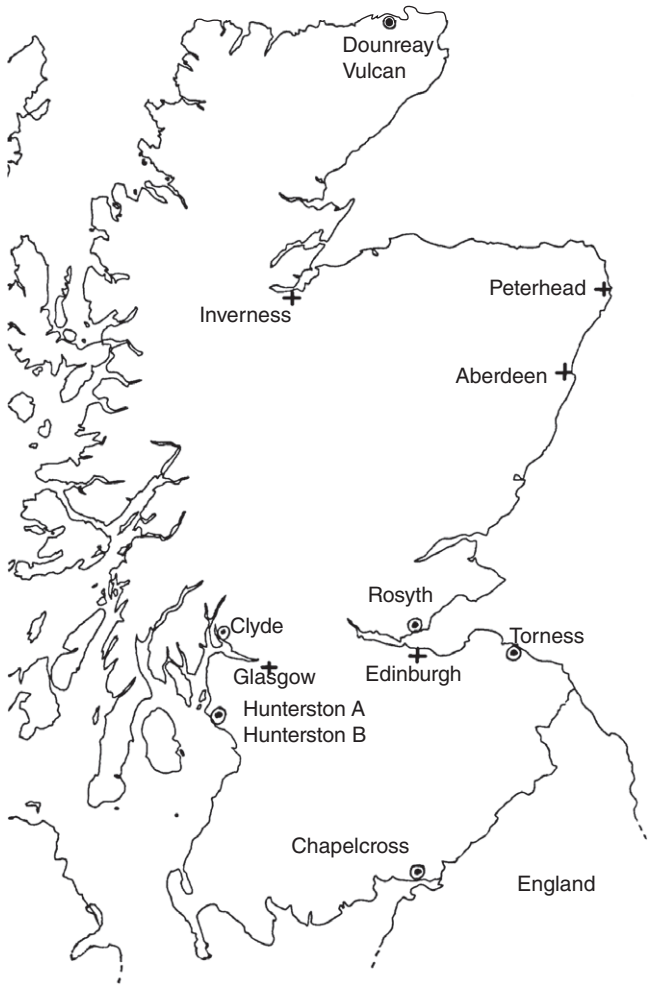
Source: Information compiled by permission of Scottish Government from Copyright data of Scottish Government in Scottish Government report B4435313 version 01 January 2011.

(UKRWI) (NDA, 2011a). Permission has been given by Scottish government for use of its copyright source data in all its higher activity waste policy documents. Permission has also been given by Dounreay Site Restoration Ltd (DSRL) and Magnox for use of its copyright source data on its sites. The MoD and Rolls Royce have both freely provided information for the sections on Rosyth Royal Dockyard and the Naval Reactor Test Establishment (Vulcan).

17.2 Legal framework for decommissioning and radioactive waste (RAW) arrangements in Scotland

17.2.1 Devolution of UK governmental powers

The Scotland Act received Royal Assent on 19 November 1998 and the Scottish Parliament sat for the first time on 6 May 1999 (White and Yonwin,



17.1 Scottish nuclear sites.

2004). This act transferred powers for specific issues to the Scottish government and reserved powers with the UK government for the remainder. The devolution of powers to the Scottish government for specific issues and not others has an influence on the areas of nuclear power, RAW management and the environment.

Among the reserved powers, the UK government controls energy, which includes electricity and nuclear energy, and also defence and national security. Among the devolved powers, the Scottish Parliament has health, planning and the environment. This means that nuclear installations in Scotland are subject to legislation in specific areas from the UK government and in other areas from the Scottish government. They are regulated by some

agencies which report to the UK government and by others that report to the Scottish government.

17.2.2 Office of Nuclear Regulation (ONR) and Scottish Environment Protection Agency (SEPA)

Nuclear licensed sites in Scotland have to comply with the terms of their site licences which are granted by the ONR which reports to the UK government. There is uniformity of approach to licensing and regulation across all UK licensed sites. ONR has responsibility for regulation of nuclear safety (Nuclear Installations Inspectorate, NII), nuclear materials transport (Department for Transport, DfT) and security (Office for Civil Nuclear Security, OCNS). On the particular subject of radioactive waste, ONR is the lead regulator for the topics of management strategy, accumulations of waste, treatment, transport and storage. The implementation of the waste hierarchy, authorisations for disposals and movements of nuclear materials are, however, regulated by SEPA which reports to the Scottish government. The waste hierarchy requires all waste managers to consider managing waste by prevention, re-use, recycling, other recovery and disposal in that order of preference. In practice, ONR and SEPA have memoranda of understanding for identifying areas where both may have an interest and for identifying which regulator will assume lead regulator status.

17.2.3 Planning

Building development on nuclear sites in Scotland is controlled by The Town and Country Planning Act (Scotland) 1997 Chapter 8 (UK Government, 1997) and The Planning etc. (Scotland) Act 2006 (UK Government, 2006). Local councils produce planning frameworks for their strategy for building and economic developments in their geographical areas. These may contain policy statements expressing a council's view on nuclear facilities or operations in their areas which may have an influence on the course of planning applications.

There is also the Scottish Councils Committee on Radioactive Substances (SCCORS), which provides a forum for discussion among those councils with nuclear interests and which can respond to nuclear issues and consultations on a joint basis.

17.2.4 Nuclear Decommissioning Authority (NDA)

The NDA reports to the UK government and its work is sponsored by the UK government and the devolved administrations of Scotland, Wales and Northern Ireland. It owns the nuclear sites and installations that are undergoing decommissioning and the low level waste repository (LLWR) near

Drigg in Cumbria. In Scotland it owns Dounreay, Hunterston A and Chapelcross sites. The NDA produces a UK-wide strategy for its whole estate (NDA, 2011b). The strategies for decommissioning the Scottish sites and the associated RAW management arrangements are agreed with the Scottish government.

All decommissioning and radioactive waste management at NDA's Scottish sites is funded by the NDA which is financed by the UK government. Justifications and business cases for expenditure of public funds on NDA's Scottish activities have to be made in accordance with UK Government Treasury rules and procedures (UK Government, 2003).

17.3 Scottish government solid low level radioactive waste (LLW) policy

The Scottish government is a joint sponsor with the UK government and other devolved administrations of the UK-wide policy for the management of LLW published in 2007 (UK Government, 2007). This policy was implemented by an enabling strategy developed by NDA in 2010 (UK Government, 2010) of which the Scottish government was again a joint sponsor. Consequently there is no difference in approach to LLW management in Scotland than anywhere else in the UK, and LLW generated in Scotland is routinely transported to the LLWR in Cumbria for disposal. The exception is LLW generated at Dounreay which is disposed of on-site and described in detail in Section 17.6.

17.3.1 Liquid and gaseous low level radioactive waste

Low level liquid effluent (LLLE) and gaseous low level waste are discharged from nuclear installations in Scotland into the environment within volumetric and radioactivity limits which are specified in authorisations that are granted by SEPA. The site operators are required to manage discharges to be as low as reasonably practicable (ALARP). They are required to produce implementation control documents that demonstrate best practicable means (BPM) and best available technology (BAT). SEPA regulates the site operators against both the authorisations and the implementation control documents. SEPA's policy is to encourage nuclear site operators to drive down liquid and gaseous discharges to the absolute minimum. In some cases this involves transferring the radioactivity from liquid and gaseous effluents to a solid medium to create a disposable LLW.

17.3.2 Amount of LLW in Scotland

The amount of LLW existing at present in Scotland and that which is estimated to arise during the operational and decommissioning lifetimes of the

Table 17.2 Low level radioactive waste in Scotland by owner and location

Owner and location	Current stored (m^3)	Total lifetime packaged (m^3)	Lifetime packaged (%)
NDA			
Dounreay	9,360	113,000	30
Hunterston A	949	57,600	15
Chapelcross	20,000	167,000	45
Total NDA	*30,309*	*337,600*	*90*
EDF			
Hunterston B	100	15,300	4
Torness	92	20,300	5
Total EDF	*192*	*35,600*	*9*
MoD			
NRTE Vulcan	0	36	<0.1
Rosyth Royal Dockyard	27	183	<0.1
HMNB Clyde	7	770	0.2
Total MoD	*34*	*989*	*<1*
Grand Totals	30,535	374,189	

Source: Figures compiled by permission of NDA and DECC from Copyright data of NDA and DECC in NDA UKRWI 2010.

nuclear facilities is given in Table 17.2. This amount of LLW is around 6% of the UK predicted total of LLW to arise (NDA, 2011a). The LLW volume estimated to arise and require disposal is dominated by the decommissioning activities on the NDA estate, being 90% of the Scottish total. A large percentage of the LLW arising from NDA sites' decommissioning activities, and that which will arise when the operating nuclear power stations undergo decommissioning, is in the form of concrete, rubble and lightly contaminated metals. This often has radioactivity levels significantly below the maximum limits for LLW of 4 GBq/te alpha and 12 GBq/te beta/gamma. There is opportunity for decommissioning sites to explore routes other than to LLWR for disposal of this type of LLW rather than using up valuable space in the LLWR.

LLW of similarly low radioactivity, but generated in very small quantities from hospitals and universities is termed very low level waste (VLLW). It is able to be disposed of safely along with municipal waste.

17.4 Scottish government higher activity waste (HAW) policy

The Committee on Radioactive Waste Management (CoRWM) produced its recommendations to the UK government and devolved administrations on the long-term management of HAW in November 2006 (CoRWM, 2006).

At that time, as a sponsor of CoRWM, the Scottish government was content with CoRWM's recommendations. The main recommendation was that geological disposal was the best available approach for the long-term management of HAW. One of the qualifying conditions was the need for robust interim storage until geological disposal could be implemented, which could take up to 40 years.

The Scottish government, however, changed its view in June 2007 when Richard Lochhead, Cabinet Secretary for Rural Affairs and the Environment, announced that the Scottish government's policy for the long-term management of HAW was to support long-term, near-surface, near-site storage so that waste is monitorable and retrievable and the need for transporting the waste is minimal (Scottish Government, 2010a).

Consequently, the framework for implementing geological disposal, Managing Radioactive Waste Safely (MRWS), published as a White Paper in June 2008 (UK Government, 2008) was not sponsored by the Scottish government. The White Paper noted that the Scottish government supported long-term interim storage and a programme of research and development.

Thereafter, the Scottish government embarked on a process (CoRWM, 2011) to develop a more detailed statement of its own HAW policy which included significant stakeholder engagement. From January to May 2010 the Scottish government consulted with the public and stakeholders on a draft Detailed Statement of Policy for Scotland's HAW (Scottish Government, 2010a) and supporting documents which comprised a Supplementary Information report (Scottish Government, 2010b) and an Environmental report (Scottish Government, 2010c).

The Scottish government published its Policy (2011a), Summary of Comments (2011b) and Post Adoption Strategic Environmental Assessment Statement (2011c) on 20 January 2011. An additional seven reports provided supporting information.

The policy sets out in detail the Scottish government's position on a wide range of HAW issues. In summary the key points are:

- The policy is enabling to allow waste managers, regulators, facility owners and the NDA to take decisions on the long-term management of HAW.
- The policy is not prescriptive and it is the responsibility of HAW managers to decide on HAW management methods on a case-by-case basis in accordance with the policy framework.
- An implementation strategy for the policy will be developed by the Scottish government.
- Long-term storage is the primary long-term management option.
- The waste hierarchy should be applied and HAW can be managed by treatment, storage or disposal.

- When disposal is employed, the facilities should be near surface (no more than a few tens of metres below surface) and near to the site where the HAW is generated.
- Disposed HAW must be able to be monitored and retrievable.
- The policy will be reviewed every ten years to assess whether technologies have developed sufficiently to warrant a change to the policy.

The policy for HAW in Scotland now differs from the rest of the UK. The Scottish government has rejected at present the concept of deep geological disposal for HAW that cannot be disposed of in near-surface facilities. The Scottish government is not taking part in the MRWS process to identify a site for, and develop, a geological disposal facility (GDF). The policy does not contain the concept of volunteerism by communities for HAW facilities which is fundamental in the MRWS process.

Operators with HAW at facilities in Scotland which is unsuitable for near-surface disposal and who previously had planned for disposal of that HAW in the projected UK GDF now need to plan for new stores and longer storage periods in the absence of an identifiable final end-point.

The generally accepted understanding of disposal is that there is no intention to retrieve the waste. In practice, this means specific retrieval features are not included in the disposal facility design. Also monitoring is applied to the surrounding environment rather than the waste itself. In the Scottish context, while there may be no intention to retrieve, the policy requires that HAW in near-surface facilities must be able to be monitored and retrievable. This introduces additional requirements for designers and operators of Scottish HAW disposal facilities for monitoring of the waste itself in the facility and for including features to enable retrieval of the waste.

17.4.1 Amount of HAW in Scotland

Information on the HAW in Scotland is given in one of the seven supporting documents to the policy, *Higher Activity Radioactive Waste in Scotland* (Scottish Government, 2011d). This document is based on the NDA 2007 UK Radioactive Waste Inventory (UKRWI) and gives detailed breakdowns of the HAW by location, type of materials, radioactivity levels and volumes. It gives information on the current amounts of HAW in store and the final lifetime packaged volumes after decommissioning has been completed. The most up-to-date inventory for the UK is now the NDA 2010 UKRWI (NDA, 2011a) and an overview of NDA higher activity waste (NDA, 2012). The newer information for Scotland is not significantly different in overall terms from the 2007 UKRWI. There is no comparable analysis of the 2010 UKRWI data in the format of the Scottish government's supporting document. For a specific review of a topic to be as accurate as possible, all three sources should be consulted. As these data are compiled from estimates

that have different levels of robustness, there is naturally uncertainty in the figures but this does not affect description of the bigger picture and developing strategies.

A summary of the HAW, by owner and location, currently stored in Scotland and the estimated final lifetime packaged volumes to around 2125 is given in Table 17.3. A summary of the HAW, by type of waste, on the same basis is given in Table 17.4. The volume of HAW currently stored and the estimated final lifetime packaged volume are both about 8.5% of the total UK HAW inventory.

The radioactivity content of the HAW is currently around 700,000 TBq and is calculated to decay to 50,000 TBq by 2150. This radioactivity content is currently around 18% of the UK total radioactivity in intermediate level radioactive waste (ILW), dropping to around 9% by 2150. Around 40% of the radioactivity content in HAW in Scotland is in ILW at Dounreay both currently and in 2150. There is no high level radioactive waste (HLW) in Scotland.

17.5 Nuclear power plants in Scotland

17.5.1 Operating civil nuclear power stations

There are two operating nuclear power stations in Scotland, Hunterston B and Torness. Both are of the advanced gas-cooled reactor (AGR) type with

Table 17.3 Higher activity radioactive waste in Scotland by owner and location

Owner and location	Current stored (m^3)	Total lifetime packaged (m^3)	Lifetime packaged (%)
NDA			
Dounreay	4,090	11,300	27
Hunterston A	2,600	8,350	20
Chapelcross	310	6,230	15
Total NDA	*7,000*	*25,880*	*62*
EDF			
Hunterston B	839	8,030	19
Torness	221	7,260	18
Total EDF	*1,060*	*15,290*	*37*
MoD			
NRTE Vulcan	9	156	0.4
Rosyth Royal Dockyard	17	116	0.3
Total MoD	*26*	*272*	*0.7*
Grand Totals	8,086	41,442	

Source: Figures compiled by permission of NDA and DECC from Copyright data of NDA and DECC in NDA UKRWI 2010.

Table 17.4 Higher activity radioactive waste in Scotland by material

Type of HAW	Current stored (m^3)	%	Total lifetime packaged (m^3)	%
Desiccant	246	3	1,398	3.0
Fuel debris	574	7	932	2.0
Pu-contaminated materials	902	11	699	1.5
Ion exchange resins	82	1	699	1.5
Other	328	4	1,165	2.5
Thorium	82	1	0	0
Graphite – short-lived	1,476	18	4,194	9.0
Activated metals	738	9	6,990	15.0
Contaminated metals	1,148	14	6,291	13.5
Raffinates	1,968	24	3,728	8.0
Sludges – short-lived	164	2	699	1.5
Sludges – long-lived	492	6	1,398	3.0
Concrete	0	0	1,165	2.5
Graphite – long-lived	0	0	17,242	37.0
Total	8,200		46,600	

Source: Figures compiled by permission of Scottish Government from Copyright data of Scottish Government in Scottish Government report B4435313 version 01 January 2011 and by permission of NDA and DECC from Copyright data of NDA and DECC in NDA UKRWI 2007 (now updated in NDA UKRWI 2010).

two reactors each, and both are currently owned and operated by EDF Energy. Together they produce around 30% of electricity generated in Scotland (Scottish Government, 2012). They both have water-filled ponds for storing spent fuel from their reactors to allow cooling of the fuel before it is transported to Sellafield for storage and reprocessing, or long-term storage. The NDA's strategy for management of spent AGR fuel is currently under review.

Hunterston B

Construction of this 1,200 MW station started in 1968 and it was commissioned in 1976. It is situated near West Kilbride in Ayrshire (Fig 17.1). It is currently scheduled to operate until 2016 (EDF Energy, 2010), although lifetime extension is being considered. Boiler restrictions currently limit it to producing around 890 MW. Normal operations generate LLW from monitoring and maintenance activities in the form of plastics, paper and scrap metals. This LLW is drummed and sent to the LLWR for disposal.

Three types of operational ILW are produced (CoRWM, 2010):

- wet wastes of organic resin, sludge and sand arising from filtration and treatment of liquid effluent to allow authorised discharge of LLLE to sea (lifetime volume ~120 m^3)

- desiccant and catalyst wastes arising from the removal of moisture from the reactor coolant gas (carbon dioxide) (lifetime volume ~640 m^3)
- activated components and fuel stringer debris arising from refuelling operations (lifetime volume ~1040 m^3).

This ILW is stored in vaults that were part of the original reactor design and construction, but there are few features incorporated to ease the eventual necessary retrieval of the waste or decommissioning of the vaults.

Decommissioning strategy is to defuel the reactors and prepare for an extended period of care and maintenance immediately after the power station finally shuts down. This would take around eight years and would include retrieval and packaging of the stored ILW. Extensive additional shielded access and retrieval and immobilisation facilities will need to be constructed to enable these decommissioning operations to be undertaken. Also a new purpose-built on-site store with a design life of at least 100 years will be required to store the conditioned ILW until the final end-point for it is identified. As an alternative, there is the possibility of utilising spare capacity in Hunterston A's ILW store (see Section 17.5.2) instead of constructing a new store, and discussions with stakeholders are being undertaken.

Depending on when the station ceases to operate, the station would enter a care and maintenance regime around the mid to late 2020s. The strategy is to continue a long care and maintenance regime of the secure and contained station to allow radioactive decay to levels where in the future the final decommissioning and demolition can be undertaken by more conventional methods with less requirement for radiological protection and remote handling methods. Consequently this period is envisaged to continue until the early 2100s. Final decommissioning and demolition in the first decades of the 2100s will create around another 6,000 m^3 of packaged ILW, mainly in the form of immobilised graphite, for storage. A further 15,000 m^3 of packaged LLW will require disposal.

Torness

Construction of this 1,230 MW station started in 1980 and it was commissioned in 1988. It is situated near Dunbar in East Lothian (Fig 17.1) It is currently scheduled to operate until 2023 (EDF Energy, 2010).

Its operational waste management processes and also its decommissioning strategy are the same as described for Hunterston B. Defuelling and preparation for care and maintenance is planned to be undertaken by around the early 2030s with the long-term secure care and maintenance period then starting and continuing until around 2110. The decommissioning waste management strategy is the same as Hunterston B but the estimated LLW that would be produced is higher and the ILW lower than for

Hunterston B (see Tables 17.2 and 17.3). Advances in the design of Torness, the later station, enabled the generation of ILW to be limited.

17.5.2 Nuclear power stations under decommissioning

Chapelcross

Construction of this 200 MW four-reactor Magnox station, the first commercial station in Scotland, started in 1955 and it was commissioned in 1959. It is situated near Annan in Dumfries and Galloway (Fig 17.1). It was shut down in 2004 and operated until 2005 by British Nuclear Fuels Ltd. It was then taken into the ownership of the NDA and is currently undergoing decommissioning on contract by Magnox, a wholly owned company of EnergySolutions Inc. (Magnox, 2012a).

Defuelling and preparation for care and maintenance is underway and is estimated to be complete by 2017. Defuelling is the major priority which started in 2008 and is planned to be complete in 2013. This is a key date because the fuel from Chapelcross is required to be at Sellafield for reprocessing through the Magnox reprocessing plant before that facility is closed and decommissioned.

Owing to its links with historical operations at Sellafield, Chapelcross has a wide range of operational ILW consisting of sludges, inorganic ion exchange resins, ceramic pellets, tritiated oils, desiccants and miscellaneous activated components. This waste is stored in ponds and vaults which were not originally designed or constructed with decommissioning in mind.

Magnox has developed strategies for the management of the accumulated operational ILW streams that are to be dealt with during preparation for care and maintenance (Magnox, 2011). The strategies for those ILW streams that will only be dealt with at final site clearance are specifically left undefined at present.

The strategy for the majority of ILW waste streams is to retrieve the ILW, condition it and then package it in high integrity containers that are designed to provide the required shielding. They can be stored in conventionally constructed unshielded buildings. There is some ILW that is associated with past arrangements with Sellafield and this will be returned to Sellafield for treatment. The target date for achieving retrieval and storage of ILW that is not destined to be dealt with at final site clearance is 2016.

Chapelcross has also made the first shipments of metal LLW to the LLWR recycling routes. Decommissioning strategy is to put the station into a long-term care and maintenance regime in 2018 to allow radioactive decay to levels where in the future the final decommissioning and demolition can be undertaken by more conventional methods with less requirement for

radiological protection and remote handling methods. This period is planned to continue to around 2090 after which the final decommissioning and demolition would take place up to 2095. LLLE is discharged to sea through a 6 km pipeline to the Solway Firth.

The final packaged volume of HAW is estimated to be around 5,000 m^3 and will be stored until a final end-point is identified. Around 75% of this HAW will be ILW graphite (NDA, 2012). Final site clearance will create around a further 30,000 m^3 of packaged LLW for disposal.

Hunterston A

Construction of this 360 MW two-reactor Magnox station started in 1957 and it was commissioned in 1964. It is situated near West Kilbride in Ayrshire on an adjoining site with Hunterston B nuclear power station. From construction, through its operating period until it was shut down in 1990, it was operated by the Southern Scotland Electricity Board (SSEB). From shut down it was managed by Scottish Nuclear and then Magnox – Electric until it was taken into the ownership of the NDA in 2005. It is currently undergoing decommissioning on contract by Magnox. At the time of opening it was Scotland's first civil nuclear generating station and the largest in operation anywhere in the world (Magnox, 2012b).

Defuelling started in August 1990 and was completed in January 1995. Currently, activities are being carried out to prepare the site for entry into the long-term secure care and maintenance period that is scheduled to start in 2022. This care and maintenance period is planned to continue to around 2080 after which the final decommissioning and demolition will take place up to around 2090.

The decommissioning strategy is the same as for Chapelcross, and all Magnox stations, but the key difference is that a shielded ILW store has been constructed on-site to store the conditioned operational and decommissioning waste in unshielded containers. These containers will be the industry standard 3 m^3 boxes and the store was designed to accommodate 1,600 containers. Further work has been undertaken on crane guidance systems and container stacking and spacing to allow the number of containers able to be stored to increase to around 2,500.

There are five vaults containing solid ILW from previous operations which were not designed or constructed for ease of eventual waste retrieval or decommissioning. A project, the solid active waste building retrieval (SAWBR) facility, was completed in 2011 to provide the means to allow entry into these vaults using remote operated machinery. The ILW will then be retrieved using remote controlled robots. The SAWBR facility includes equipment for packaging the retrieved waste into 3 m^3 boxes so that it can be transported across site to an encapsulation plant.

A retrieval and packaging plant for the wet ILW is under construction above the wet ILW storage vault. This facility is of the 'canyon' design which, though it is a shielded facility, it nevertheless has an open top to allow different equipment to be installed or removed depending on the processing required (CoRWM, 2010). The transport system and encapsulation plant for both solid and wet ILW are in the construction stage and are planned to be commissioned in 2013.

The final packaged volume of HAW is estimated to be around 8,400 m^3 and will be stored until a final end-point is identified. Around 51% of this HAW will be ILW graphite and 32% fuel element debris (NDA, 2012). Around 58,000 m^3 of packaged LLW will require disposal.

17.6 Dounreay research station under decommissioning

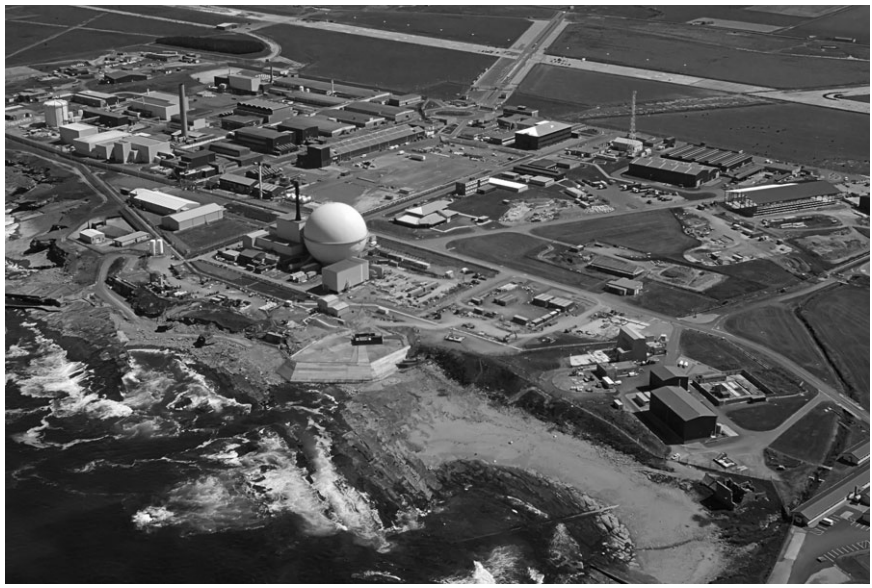
The UK government started construction of the Dounreay research station near Thurso in Caithness in 1955 to undertake a major research programme on fast reactor technology (Fig 17.1). The programme was stopped in the mid-1990s as it was considered that fast reactors were not needed in the foreseeable future. The research station was operated by the United Kingdom Atomic Energy Authority (UKAEA) until 2005 when it was taken into ownership by NDA and is currently managed under contract by Dounreay Site Restoration Ltd (DSRL) a wholly-owned subsidiary of Babcock Dounreay Partnership. The area of the licensed site is 57 ha situated within 547 ha of NDA-owned land.

During the 50 years of operations, three large nuclear reactors were built, two fast reactors and a materials research reactor. Each had associated industrial size fuel research and inspection facilities, associated fuel reprocessing facilities and RAW management facilities (Dounreay, 2012). Both fast reactors also had their own steam generating plant of unique designs that drove a conventional electricity generating plant. The scale of the installations can be seen in Fig. 17.2.

Owing to the wide range of research and the goal of demonstrating the complete fast reactor fuel cycle, which was achieved in the early 1980s, Dounreay has the widest range of radioactive wastes and facilities to manage and the most complex decommissioning challenges of any nuclear site in Scotland. In a UK context, only Sellafield in England has a more hazardous and complex RAW and decommissioning programme.

17.6.1 Dounreay materials test reactor (DMTR)

Construction was started in 1955 and the DMTR was the first nuclear reactor to operate in Scotland. Its purpose was to test materials under



17.2 Dounreay fast reactor research site. Photograph courtesy of DSRL.

nuclear radiation conditions for the future fast reactor programme. It produced 25 MW thermal but not electricity and was shut down in 1969.

Early decommissioning after shutdown was limited to post-operational clean out (POCO) and removal of non-active support facilities. Decommissioning during the early 2000s has been more comprehensive with cleaning out of the fuel pond and cells and the demolition of inactive buildings. DMTR is now in a care and maintenance period prior to final decommissioning and demolition programmed to start in 2015.

17.6.2 Dounreay fast reactor (DFR)

This is the iconic steel sphere on the north coast of Scotland. It was built between 1955 and 1958 and was the first nuclear reactor in the world to supply electricity to a national grid in 1961. It was cooled with a eutectic mixture of sodium and potassium (NaK) and produced 14 MW. It was shut down in 1977.

Early decommissioning included removing the conventional electrical generation installations, removal of the fuel and destruction of the secondary circuit NaK. There was then a lengthy period of care and maintenance during which numerous theoretical studies and large off-site and on-site practical experiments were carried out to investigate a way in which to decommission the reactor. The major challenges were how to remove, treat and destroy the

57 tonnes of highly contaminated (400TBq) primary circuit NaK and then remove the uranium and plutonium breeder fuel elements that had provided the blanket around the core. Both challenges are among the most hazardous and difficult in NDA's UK decommissioning programme.

During the period 1999–2008, the original plant used to destroy the primary NaK was refurbished and revalidated along with major refurbishment of the DFR infrastructure. A successful campaign of destroying the NaK started in 2009 and is nearing completion. The process involves reacting the NaK with an aqueous solution of NaOH in a comprehensively monitored and controlled reaction vessel and then neutralisation of the aqueous waste stream produced with HCl. The radioactive caesium is removed from this waste stream by ion exchange plant using hexacyanoferrate inorganic resin. The decontaminated salty water is discharged to sea through the site's low level liquid effluent treatment plant (LLLETP). The contaminated ion exchange medium is then stored as solid ILW.

The internal surfaces of the reactor and the 9 km of heat exchanger circuit pipework require the residual NaK to be removed. A tightly controlled wash out process utilising water vapour and nitrogen, or a dry process associated with dismantling could be possible methods.

Special equipment has been designed and manufactured to cut out the remaining uranium and plutonium breeder elements. When removed, they will be treated and packaged in the specifically designed shielded cells that have been constructed as a new facility abutting the sphere. The packaged breeder fuel will then be transferred to Sellafield for reprocessing.

There is then a programme of decommissioning the pipework, NaK processing vessels and the reactor vessel itself in the period up to the 2020s. The current strategy is for the sphere itself to be demolished unless a future viable initiative emerges for it to be retained as a historical industrial heritage monument.

17.6.3 Prototype fast reactor (PFR)

This was the last research fast reactor in the research programme. The next reactor would have been a commercial size demonstration fast reactor. Construction of PFR started in 1968 and it operated from 1974 to 1994 with an output of 250 MW. Although the reactor was not of commercial size, the PFR fuel assemblies were designed and manufactured at commercial size. It was cooled by sodium as enough experience and confidence had been gained from DFR operations to go forward without the need for a eutectic coolant.

After shutdown, the fuel was removed to the associated fuel pond where it remains until a decision on its final treatment and destination is made.

The experience gained from the early DFR NaK decommissioning enabled the world's largest sodium destruction plant to be designed and

built during the late 1990s. The plant was built in the decommissioned and stripped out turbine hall. Around 1,500 tonnes of primary and secondary sodium were destroyed by the same process as that used for DFR. The difference from DFR was that the sodium was only lightly contaminated as PFR fuel was in sealed elements whereas DFR fuel was in vented elements so reaction products contaminated the NaK.

As with DFR, there is a current programme for cleaning up the residual sodium and decommissioning the pipework, vessels and reactor components over the next decade. The additional challenge of decommissioning the PFR reactor itself is that it is 15 m below ground level and this requires the design and operation of special remote handling appliances.

Decisions in the UK and Scotland within the nuclear industry and with regulators on the concepts and practicability of *in-situ* disposal will have a bearing on the eventual end state of the decommissioned PFR reactor.

17.6.4 Radioactive waste management facilities

From the start of active operations at Dounreay, the site has managed its radioactive waste in its own waste management facilities.

LLW

Alone amongst UK nuclear sites, it has disposed of its LLW to near-surface disposal pits on site instead of sending the packaged LLW to the LLWR. A series of six disposal pits were constructed and operated from 1959 until 2005. These are situated at the north end of the site adjacent to the sea and contain around 33,000 m^3 LLW. Pits 1–4 are unlined and accepted tumble tipped bagged LLW. Pits 5 and 6 have concrete bases and the LLW was disposed of in uncompacted or supercompacted 200 litre drums. All the pits contain large bulk items and are now capped off with rock and soil. Water ingress to the pits is collected in sumps and pumped to the LLLE treatment plant.

During the 1990s, it was clear that further extension of the existing LLW disposal facility was impracticable and would not meet current environmental regulatory requirements. In 1999, a best practicable environmental option (BPEO) study (UKAEA, 2004a) was initiated to determine the most appropriate way in which to manage the continuing operational LLW and LLW that would be generated from decommissioning the whole site (approximately 150,000 m^3 of packaged LLW). The BPEO study included significant stakeholder involvement (Broughton and Tait, 2008; Broughton, 2003). Preliminary performance assessment work and environmental impact studies (UKAEA, 2004b) were carried in parallel to inform the BPEO study. Eventually this led to the BPEO being identified as the construction

of six new disposal vaults on the Dounreay estate south east and inland from the licensed site boundary. Their positioning was influenced by 10,000-year sea level rise assumptions. The BPEO proposal was endorsed by the Scottish Government Environment Minister in May 2005 (Scottish Executive, 2005) and the planning application for this development was granted by the Highland Council in January 2009 (Highland Council, 2009). The project to construct the first two of the new vaults and associated facilities is currently in progress with the target of bringing them into operation in 2014. The vaults are of concrete construction with steel roofs and extend 15 m below ground level. Comprehensive water management features are included. The LLW will be disposed of in grouted half-height ISO containers (HHISO) placed in one of the new vaults. Decommissioning waste of very low activity, but high volume (DLLW) such as concrete, rubble and steel will be disposed of in the other new vault in a designated bulk manner.

LLLE

LLLE is a continuous product from current decommissioning and current waste management operations. The LLLE is discharged to sea through a system that was refurbished in 1992 and a LLLE treatment plant (LLLETP) that was brought into operation in 1997 to replace the original one. All discharges have been, and are, authorised by SEPA.

ILW

ILW has been produced from maintenance, waste management and reprocessing activities associated with the fast reactor research programme. ILW is currently being produced from decommissioning of the facilities. As the fast reactor fuels contained plutonium, there is significant alpha contamination associated with many of Dounreay ILW streams. This leads to specific handling and containment requirements in all waste management and decommissioning operations.

The Dounreay shaft and wet silo

During the construction of the tunnel for the LLLE pipework in the 1950s, a vertical access shaft was sunk near the shoreline. After construction of the tunnel, the access shaft was sealed off from the tunnel with an *in-situ* cast concrete plug. In 1958 this 4.6 m diameter, 65 m deep shaft was authorised by the UK government agency of the time to allow its use as a disposal facility for ILW. A wide variety of ILW, ranging from soft waste in bags to complete steel machinery, was tumble tipped into the shaft until 1977 when an explosion in the shaft led to the cessation of disposals.

The shaft is unlined and so there is groundwater ingress with the result that the water residing in the shaft becomes contaminated. The water level in the shaft is kept below that of the surrounding groundwater level by pumping. The extracted water is discharged through the LLLE authorised route.

During the late 1960s UKAEA recognised that the tumble tipping of ILW into a near-surface facility that was not designed as a disposal facility and had no engineered barriers between the ILW and the host environment would become an unacceptable practice. A replacement vault, the wet silo, was constructed as a single skin tanked concrete walled vault excavated into the host rock. It came into operation as a store in 1971. Its waste entry ports are at ground level and ILW was dropped into the silo from bottom opening vertical flasks through shielded gate valves. This facility is water filled to aid cooling and to provide shielding of the ILW.

Around 1980 UKAEA recognised that the historical disposals to the shaft, and emplacements in the wet silo had been so divorced from current practices that the ILW should be retrieved from these facilities. There had been no thought of ever retrieving ILW from the shaft but it had always been planned to empty the wet silo as it was licensed as a store. However, neither facility has any features to enable emptying and decommissioning and both present major challenges. Engineering studies and practical experiments for waste treatment have been carried out to inform the future retrieval projects.

As there are no in-built monitoring features to detect possible leakage in either facility, a number of boreholes have been sunk over the years in the shaft and wet silo areas. Monitoring of the groundwater in these boreholes has not shown increases in the levels of radioactivity to be of significance to the wellbeing of the environment or operators working on the site (Environment Agency *et al.*, 2010).

In preparation for retrieval of ILW from the shaft, a major project costing £27 million was completed in 2008 to encircle the shaft with a series of boreholes through which grout was pumped to form a water barrier around and under the shaft. This barrier is not completely impervious but has reduced the groundwater flow into the shaft by a factor of 10–15. Although not a design parameter, the grout curtain provides additional retention for any radioactivity migration from the shaft. The reduced ingress of water into the shaft will allow practicable contaminated water management to be undertaken when retrieval of the ILW is underway.

Miscellaneous ILW storage

In parallel with running down and eventual cessation in 1998 of emplacement of ILW into the wet silo, an above ground ILW store was brought into

operation that allows retrievable storage of ILW in 200 litre drums in shielded vertical channels. Emplacement is carried out by conventional transport flasking operations.

Plutonium-contaminated material (PCM) storage

Waste is generated that has the gamma and beta radioactivity levels in the LLW range but is alpha contaminated. It is contact handleable with precautions, and referred to as PCM. There is an operational above ground vault store which accommodates the PCM in 200 litre mild steel drums awaiting further treatment and a final end-point being identified.

ILW treatment and storage associated with research reactor fuel reprocessing

Reprocessing of fuel from DMTR produced ILW waste liquors called raffinates that have been stored in underground tanks housed in stainless steel lined shielded vaults. In the late 1980s and early 1990s, the Dounreay cementation plant (DCP) was constructed. This plant takes the raffinate from the storage tanks in batches and then mixes measured quantities of raffinate and cement powders in 500 litre stainless steel drums to form a monolithic solid wasteform that can be stored for at least 100 years prior to a final end point being identified. This wasteform has a Letter of Compliance (LoC) from the Radioactive Waste Management Directorate (RWMD) of NDA accepting its suitability for geological disposal.

A dedicated ILW vault store has been constructed for storing these 500 litre drums. It can also store overpacked 200 litre drums retrieved from the miscellaneous ILW store. It has an import/export facility to allow transfer of the drums in transport flasks to other facilities when the need arises.

ILW treatment and storage associated with fast reactor fuel reprocessing

Raffinate from reprocessing DFR and PFR fuel is also stored in dedicated tanks in the same underground lined and shielded vaults as the research fuel raffinate. In this case, though, there is not yet a facility at Dounreay to treat and encapsulate this particular raffinate.

Gaseous LLW

All operational facilities at Dounreay have general space extraction and fresh air ingress ventilation systems and, if necessary, dedicated systems for contained active equipment ventilation. The latter is usually associated with gloveboxes, shielded cells and transfer systems. The extracted air is filtered

and monitored before discharge to atmosphere under an authorisation from SEPA. There are a number of individual discharge stacks for specific facilities, but the fuel cycle area is served by an integrated system installed in 2010 to provide active ventilation requirements for both the current operations and the decommissioning of the complex and interconnected fuel cycle plants and laboratories (IMechE, 2010).

17.6.5 Contaminated land

There are areas of land at Dounreay associated with historical operations and accidents that have radioactive contamination above background levels but which do not constitute a significant risk to the environment or workers on the site. There is both UK- and Scotland-wide discussion on the approach to remediation of such land including the consideration of treating it as *in-situ* disposal.

17.6.6 Unauthorised discharges of radioactive particles

A difficult situation for previous and current owners and operators of Dounreay has been the contamination of Sandside beach neighbouring Dounreay and the immediate foreshore and seabed by radioactive particles (Rodriguez *et al.*, 2005; Rodriguez, 2009). These particles originate from the mechanical cutting operations involved with reprocessing of DMTR fuel and have activities in the range 10^3–10^8 Bq ^{137}Cs. They are thought to have been unknowingly discharged to sea through the LLLE system during the 1960s and 1970s. The discharges of the particles, although not intended, were nevertheless unauthorised. Although risk studies by UK national institutions (Harrison *et al.*, 2005) indicate there is not a significant risk to the public by the presence of these particles, there is local concern about past discharges and current finds of particles on the publicly accessible beach (208 particles from 1983 to early 2012). The outcome of long-term environmental studies, academic reviews and public consultation on how to deal with the problem has been to instigate a programme of recovery of particles in an area of 60 ha of the seabed off Dounreay. The recovery operations are carried out by specialist diving teams using remote controlled seabed vehicles.

17.6.7 Exotic fuels

A wide range of unirradiated and irradiated uranium and plutonium mixtures of fast reactor fuels has been left over from the research programme. These require a high level of security for the site and their storage arrangements. The NDA reviewed the credible options for this fuel which included

stakeholder consultation. The top two options were continued storage at Dounreay or transfer to Sellafield. The former would require rebuilding of stores over a 100-year period and continuing high level security arrangements. The latter would allow use of common facilities and security at Sellafield but would entail transfers through many communities. The decision to transfer the exotic fuels to Sellafield as the preferred option was made in February 2013 (NDA, 2013).

17.7 Nuclear submarines and naval test reactors in Scotland

17.7.1 Naval reactor test establishment (NRTE) Vulcan

Vulcan is situated in Caithness adjacent to the Dounreay site. The site is owned by the MoD on a long lease from the NDA and operated by Rolls Royce. Its purpose is to test nuclear submarine propulsion reactors on shore in support of the operating fleet.

Construction of the Dounreay Submarine Prototype 1 (DSMP1) was started in 1957 and the first reactor was operational in 1965. The facility tested a number of reactor cores until it was shut down in 1984. The facility includes a pond where fuel from the testing programme is stored.

A second facility, the shore test facility (STF) was commissioned in 1987 for a similar testing programme on the next generation of submarine reactors. It is planned to operate this facility until 2015 when it will no longer be required (UK Government, 2011) as a reactor core prototype plant. Associated with the STF is a pond where fuel from this testing programme is stored and a decontamination and waste treatment facility (DWTF) in which is stored activated organic resins from decontamination operations in the STF. Operational LLW from Vulcan is transferred to Dounreay for disposal and LLLE is transferred to the Dounreay LLLETP.

Post-operational activities and early decommissioning could start in 2015 and be completed by 2021. Options to continue support to the naval nuclear propulsion programme from the Vulcan site are being considered together with a decommissioning programme. Final decommissioning and demolition could take up until 2050 to be completed. Some of this could be planned in and associated with the decommissioning programme at Dounreay. The decommissioning waste volumes are small compared to Dounreay and could be incorporated into Dounreay's management arrangements. The lifetime packaged volume of LLW to be disposed of at Dounreay is estimated to be around 3,600 m^3. The lifetime packaged volume of ILW, possibly to be stored at Dounreay, is estimated to be around 156 m^3 (NDA, 2011a). However, the Scottish HAW policy does not apply to Vulcan so the final end-point for this ILW may be different from that of Dounreay's.

17.7.2 Her Majesty's naval base (HMNB) Rosyth Royal dockyard

Rosyth is a long established naval dockyard built between 1909 and 1915. It covers 127 ha and is located on the north side of the Firth of Forth in Fife (Fig 17.1). The dockyard became involved with nuclear operations in 1960 with the start of support services to the Royal Navy's nuclear submarines. Some support work continued until 2003 although since 1993 the main support services have been provided at Devonport in England. In 1997 the dockyard was sold to Babcock International which now holds the nuclear site licence. The nuclear decommissioning liability is still retained by the MoD.

Submarines

There are seven redundant nuclear submarines laid up floating at Rosyth and current operations are focused on one-year, six-year and twelve-year maintenance routines for each submarine to ensure they are kept in a safe state and that they will be in a condition suitable for their eventual decommissioning. The strategy for decommissioning the UK's fleet of nuclear submarines was the subject of a consultation exercise carried out in late 2011/early 2012 (MoD, 2011). No date for deciding on the chosen strategy has been made but there are two significant conditions which will affect the decision and its timing. Firstly, decommissioning will not commence until a storage solution for the ILW arising has been agreed. This is a joint MoD and NDA programme in itself. Secondly, berthing space for laying up redundant submarines will be full by 2020, so if decommissioning has not started, then further berthing facilities would be required. The current favoured option in the consultation is that the seven laid up submarines at Rosyth would be decommissioned there, but none of the submarines which are operational at present would go to Rosyth for decommissioning.

In 2000, a joint MoD and Babcock project team decided that the nuclear support facilities that would become redundant in 2003 should be decommissioned with the objective of de-licensing the nuclear site area of 0.83 ha to allow future industrial use. The first operations, which took four years, were to characterise the radioactive contamination, agree on monitoring protocols with the regulators and obtain the necessary authorisations from SEPA. For thoroughness in characterisation, retired employees were interviewed for their knowledge of historical discharges or spills, health physics logbooks were checked and the GPS-linked 'Groundhog' monitoring system was employed.

Rosyth has an active waste accumulation facility (AWAF) for storing LLW and ILW. It also has a LLLE outlet from the end of one of the dock's

piers. Monitoring of the sediments in the tidal and non-tidal basins detected no significant radioactivity.

The first phase of decommissioning and demolition of redundant facilities was undertaken by contractors and completed in 2009 with 99% recycling of non-asbestos building materials. This has led to low volumes of LLW requiring disposal at LLWR. Contaminated metals were authorised by SEPA to be sent to Studsvik AB in Sweden for treatment. 96% by weight was recyclable by Studsvik and one tonne of LLW was received back which was disposed of at the LLWR. A major facility decommissioned and demolished was the health physics building which contained the LLLE treatment plant. As LLLE treatment is a continuing requirement, a mobile unit has been procured.

The ILW waste from the decommissioning to date is organic ion exchange resin which is being stored in AWAF in 1.2 m^3 transport container tanks. The strategy agreed with regulators and LLWR is to condition these resins in cementitious grout directly in one-third height ISO containers. The monolithic wasteform is LLW which is acceptable for disposal at LLWR. On this basis, the AWAF could be closed in 2016.

The lifetime packaged LLW disposed of at LLWR is estimated to be around 183 m^3.

Decisions on delicensing are in abeyance awaiting the determination of the strategy on the SDP.

17.7.3 Her Majesty's naval base (HMNB) Clyde

Usually referred to as 'Faslane', this Royal Navy establishment is situated on Gare Loch off the Firth of Clyde in Argyll and Bute and near to Glasgow (Fig. 17.1). It was constructed in the 1940s and is now the principal support base for the UK's operational nuclear submarine fleet. The volume of operational radioactive waste produced by servicing the submarines is small compared to refitting or decommissioning work carried out at Rosyth or Devonport, and based on projected operations the lifetime packaged volume of LLW to be disposed of at LLWR is 770 m^3 (NDA, 2011a).

17.8 Industry and small users

17.8.1 Oil and gas industry

Extensive oil and gas industrial activity takes place in the North Sea to the east and north east of Scotland. The main on-shore centre of the industry in Scotland is Aberdeen. Drilling and processing operations create waste sludges and films contaminating the drilling and processing equipment. These wastes contain naturally occurring radioactive materials (NORM). Although the levels of radioactivity (20–100 Bq/gm) are usually insufficient

to cause concern to the health of workers exposed to them, or to the environment, precautions are taken to limit the build-up and accumulation of the wastes. The industry has a well-developed system of cleaning its pipe drilling strings, pipework, valves and other process equipment through the use of contractors.

The cleaning process, usually referred to as descaling, creates waste containing very low levels of radioactivity. This waste was routinely disposed of to sea near Aberdeen until 2011 when the licence was withdrawn. A facility was opened in October 2011 by a joint venture of SITA UK and Nuvia at Stoneyhill landfill site near Peterhead to process NORM. This facility cleans the NORM from the equipment by high pressure water jetting which is then encapsulated in cement in drums. The drums of cemented NORM are then disposed of under controlled conditions in the adjacent landfill facility which is operated by SITA UK (Sita, 2011).

17.8.2 Hospitals and small users of radioactive materials

Scotland is similar to the rest of the UK in its hospitals and industries using radioactive sources for medical and industrial purposes. The use of these sources is controlled by the suppliers who in most cases are also responsible for their storage or disposal after use. There are many movements of these radioactive sources daily under controlled conditions and in authorised containers.

17.9 Conclusion

Although the volumes of radioactive waste and decommissioning activities in Scotland are small compared with the total UK liabilities, they are nevertheless diverse and challenging. Dounreay is the second most challenging site in the UK after Sellafield. Scottish radioactive waste managers and nuclear site operators manage their responsibilities both within UK requirements and legislation and the Scottish government's specific policies on RAW management. Although Scotland has significantly different approaches to some aspects of HAW management, these are not creating operating problems at present. During the next few decades of development of disposal technologies in the UK as a whole, and the Scottish government's commitment to review its HAW policy every ten years, closer alignment and coordination are possible.

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18

United States: experience of radioactive waste (RAW) management and contaminated site cleanup

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Abstract: The federal government of the United States is responsible for the safe disposal of spent nuclear fuel and high-level radioactive waste. The development of policies and practices has evolved over the years to ensure that the waste is managed appropriately. The major agency involved in the implementation of these activities is the Department of Energy (DOE), and the regulatory authority is assigned to the Nuclear Regulatory Commission (NRC) and Environmental Protection Agency (EPA). The US waste classification system is divided into two areas – commercial and government owned. Current storage and disposal techniques are described, addressing the different types of waste. The cleanup history and current strategies for these waste types are discussed in detail to provide the reader with an overall understanding of the US national waste management system.

Key words: radioactive waste, regulations, Department of Energy (DOE), Nuclear Regulatory Commission (NRC), low-level waste (LLW), high-level waste (HLW), mixed waste, spent fuel, storage, disposal, transuranic (TRU) waste, uranium mines and mills, Waste Isolation Pilot Plant (WIPP), cleanup program

18.1 Introduction

The United States operates waste storage facilities for low-level waste (LLW) and transuranic (TRU) waste. It is the only country in the world that has successfully licensed, constructed, and now operates a deep geological repository for defense-generated radioactive waste (RAW), the Waste Isolation Pilot Plant (WIPP). There are three main sources of nuclear

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waste in the United States that resulted from either defense or civilian applications:

1. Legacy waste from military operations – defense waste was the first type of radioactive waste generated. It is the byproduct of nuclear weapons production. Legacy waste from defense applications includes materials of multiple compositions and forms, presenting challenges for stabilization before disposal. In general, the management of legacy waste consists of a highly integrated operation that involves storing liquid waste in underground tanks; removing, treating, and dispositioning the low-activity fraction in concrete vaults; and vitrifying and storing the higher-activity waste until permanent disposal at a federal repository. There are 88 million gallons of liquid waste stored in tanks, 1.5 million m^3 of solids, and a variety of contaminated equipment. In addition, there are surplus weapons materials and spent nuclear fuel (SNF) from reactors on naval vessels.
2. Fuel cycle operations for energy production – civilian waste that results from fuel cycle stages for electricity production. This waste is the byproduct of facilities used for:
   - uranium mining and milling – waste consists mainly of sandy tailings whose composition is the same as uranium ore (absent uranium)
   - conversion, enrichment, and fuel manufacturing – the main byproduct is depleted uranium (DU) stored as either UF_6 or U_3O_8
   - electricity generation – the main waste in terms of activity is spent fuel, which consists of highly radioactive fission products and transuranic elements, and is classified as high-level waste (HLW). Nuclear wastes resulting from these operations are stable, unlike defense legacy waste, and may be readily stored and disposed. In addition to spent fuel, other low- and intermediate-level waste is generated from support and decommissioning operations.
3. Others types of waste – research and development, accelerators, medical, industrial, and naturally occurring. This waste composition is mainly short-lived radionuclides, usually classified as LLW, and is mainly stored onsite until it decays.

Both defense and civilian applications produced radioactive waste ranging from LLW to HLW. Defense and civilian generated waste have similar characteristics in terms of radiotoxicity and need to be isolated from the public; however, their forms are significantly different and the waste conditioning necessary before disposal differs significantly:

- Defense waste is mainly characterized as radioactive material in a very diluted form, whereas civilian waste is mainly generated in a concentrated form.

- Defense waste needs to be concentrated and converted to a stable form before disposal, whereas civilian waste in the absence of reprocessing may be directly packaged, stored, and disposed.

Nuclear waste from both civilian and defense applications varies in its composition and form. In general, the nuclear waste attributes that affect humans and the environment and that determine the disposal path are chemical composition, physical form, and type of radiation. To facilitate a safe and cost effective waste disposal strategy, waste is categorized to provide guidance for its handling, transportation, storage, and ultimately final disposal. It is important to understand that how the waste is categorized ultimately affects how its final disposition is determined. The classification system ranges from very low-level waste (VLLW) to HLW. It varies from country to country but falls into two main types: those that are based on 'where' the waste was generated (i.e., point of origin) and those that are based on the 'intrinsic qualities' (i.e., risk-based parameters) of the material. The United States adopted a point of origin system, whereas the international community uses a risk-based system.

This chapter describes the current radioactive waste (RAW) management programs in the United States. The distinct policies, practices, and regulatory standards are explained, as well as the unique US waste classification system used. Strategies for implementing the RAW management programs are explained for different currently existing US facilities. Multiple US storage and disposal facilities contain various defense and commercial RAW (Fig. 18.1), which are discussed later in the chapter. The last

18.1 Dry cask storage of spent nuclear fuel (Nuclear Energy Institute).

sections address the cleanup and closure process for specific US radioactive waste facilities, and the lessons learned from past experiences.

18.2 Policies and practices

18.2.1 Radioactive waste policy

US national nuclear activities policy

The US government is responsible for the safe disposal of SNF and HLW. This section summarizes US policies and practices for SNF and radioactive waste management, and related nuclear activities.

The US government promotes the development of commercial nuclear power and nuclear technology for beneficial uses in medicine, industry, and research. The federal and regulatory duties for commercial and government sectors are assigned to different agencies, which are discussed later in this section.

Commercial sector

Owners and operators of nuclear power plants (NPPs) and other types of facilities manage the SNF and radioactive waste they generate at their facilities prior to disposal. The Department of Energy (DOE) is responsible for the disposal of SNF. US federal or state governments regulate waste disposal sites. Government custody may occur at different stages of the waste management process depending on the type of RAW and generating activity, including decommissioning activities.

Successful management of SNF and RAW requires careful integration among power or research reactors, waste generators, storage facilities, treatment facilities, and disposal sites, as well as their transportation interfaces. Integration is achieved through interface management, such as specified waste acceptance criteria. Acceptance requirements define the interfaces, allowing generators and disposers to have a common understanding of the waste. The United States recognizes the importance of this integration and manages the interfaces between various steps (e.g., storage, transportation, and disposal).

Government sector

The DOE is responsible for and performs most of the SNF and RAW management activities for government-owned and -generated waste and materials, mostly located on government-owned sites. These activities include managing SNF remaining from decades of defense reactor operations, which ceased in the early 1990s. Since then, the DOE has safely stored the

remaining defense SNF and SNF generated in a number of research and test reactors. The DOE also provides safe storage for the core of the decommissioned Fort St. Vrain gas-cooled reactor and the core of the Three Mile Island Unit 2 reactor damaged in a 1979 accident.

The DOE has a system for managing government SNF and radioactive waste. This includes numerous storage and processing facilities (treatment and conditioning), such as operating disposal facilities for LLW and TRU waste. Other waste management treatment and disposal systems support cleanup and closure of decommissioned facilities no longer serving a DOE mission.

The United States also continues activities to remove and/or secure high-risk nuclear and radiological materials both domestically and internationally. Part of this initiative is continuing the program of accepting US-origin foreign research reactor SNF and returning it to the United States for safekeeping and recovery of disused sealed sources.

18.3 Regulations and standards

18.3.1 Legislative system

The policy on regulatory control of RAW management in the United States has evolved through a series of laws establishing federal agencies responsible for the safety of radioactive materials. Federal legislation is enacted by Congress and signed into law by the President. US laws apply to all 50 states and its territories. Table 18.1 identifies key US laws governing radioactive waste management; pertinent legislation on the safety of SNF and RAW dates from the 1950s.

18.3.2 Regulatory system

The regulatory system for SNF and radioactive waste management in the United States involves several agencies: the Nuclear Regulatory Commission (NRC), regulating the commercial nuclear sector; the Environmental Protection Agency (EPA), establishing environmental standards; and the DOE, regulating government programs. Some NRC regulatory authority – excluding SNF, special nuclear material sufficient to form a critical mass, and HLW – can be delegated to the 50 states of the United States and the territories Puerto Rico and the District of Columbia under the Agreement State Program. This authority includes regulating commercial LLW disposal sites and uranium mill tailings sites, and regulatory authority over the disposal of mill tailings. Some states also have regulatory authority delegated to them by the EPA, such as for discharges from some industrial or mining practices.

Table 18.1 Key US laws governing RAW management

Atomic Energy Act of 1954, as amended, established the Atomic Energy Commission, the predecessor to the Nuclear Regulatory Commission and the Department of Energy, with federal responsibility to regulate the use of nuclear materials including the regulation of civilian nuclear reactors. Under Reorganization Plan No. 3 of 1970, which created the US Environmental Protection Agency (EPA), authority to establish generally applicable environmental standards was transferred to the EPA along with authority to provide federal guidance on radiation protection matters affecting public health.

The Price-Anderson Act (1957) was enacted to encourage development of the nuclear industry and ensure prompt and equitable compensation in the event of a nuclear incident. The Act provides a system of financial protection for persons who may be liable for, and persons who may be injured by, such an incident.

Solid Waste Disposal Act of 1965, as amended, requires environmentally sound methods for disposal of household, municipal, commercial, and industrial waste. The Resource Conservation and Recovery Act is an amendment to the Solid Waste Disposal Act.

National Environmental Policy Act of 1969, as amended, requires federal agencies to consider environmental values and factors in agency planning and decision making.

Clean Air Act of 1970 is the comprehensive federal law that regulates air emissions from stationary and mobile sources.

The Marine Protection, Research, and Sanctuaries Act of 1972, also known as the Ocean Dumping Act, prohibits the dumping of material into the ocean unreasonably degrading or endangering human health or the marine environment.

Safe Drinking Water Act of 1972, as amended, protects public health by regulating the nation's public drinking water supply; it requires actions to protect drinking water and its sources: rivers, lakes, reservoirs, springs, and groundwater wells.

Energy Reorganization Act of 1974, as amended, abolished the Atomic Energy Commission and established the Nuclear Regulatory Commission and the Energy Research and Development Administration – the predecessor of the DOE.

Resource Conservation and Recovery Act of 1976, as amended, regulates the handling and disposal of hazardous wastes, which are generated mainly by industry, also requires that open dumping of all solid wastes be brought to an end throughout the country by 1983.

Department of Energy Organization Act (1977) brought together most of the Government's energy programs, as well as defense responsibilities that included the design, construction, and testing of nuclear weapons into the new Department of Energy. The Department was established on 1 October 1977, assuming the responsibilities of the Federal Energy Administration, Energy Research and Development Administration, the Federal Power Commission, and parts and programs of several other federal agencies.

Uranium Mill Tailings and Radiation Control Act of 1978, as amended, vested the EPA with overall responsibility for establishing health and environmental cleanup standards for uranium milling sites and contaminated vicinity properties, the Nuclear Regulatory Commission with responsibility for licensing and regulating uranium production and related activities, including decommissioning, and the Department of Energy with responsibility for remediating inactive milling sites and long-term monitoring of the decommissioned sites.

Table 18.1 Continued

Comprehensive Environmental Response, Compensation, and Liability Act of 1980 as amended, also known as Superfund, provided the EPA with authority to address abandoned hazardous waste sites and outlined the process to be followed in identifying and remediating sites, including determination of cleanup levels and pursuit of contribution to the cleanup or cost recovery against parties deemed to have contributed to the contamination. It includes radionuclides as a hazardous substance.

Low-Level Radioactive Waste Policy Act of 1980 and Low-Level Radioactive Waste Policy Amendments Act of 1985 gave individual states – rather than the federal government – responsibility to provide disposal capacity for commercial Class A, B, and C low-level waste; authorized the formation of regional compacts (groups of states) for the safe disposal of such low-level waste; and allowed compacts to decide whether to exclude waste generated outside the compact. The acts gave the federal government responsibility for the disposal of greater-than-class C low-level waste that results from activities licensed by the NRC or Agreement States.

National Security and Military Applications of Nuclear Energy Authorization Act of 1980. Section 213 (a) of the Act authorizes Waste Isolation Pilot Plant 'for the express purpose of providing a research and development facility to demonstrate the safe disposal of radioactive wastes resulting from defense activities and programs of the U.S. exempted from regulation by the US Nuclear Regulatory Commission.'

West Valley Demonstration Project Act of 1980 authorized the DOE to conduct a technology demonstration project for solidifying high-level waste, disposing of waste created by the solidification, and decommissioning the facilities used in the process. The Act required the DOE to enter into an agreement with the State of New York for carrying out the project.

Nuclear Waste Policy Act of 1982 as amended by the Nuclear Waste Policy Amendments Act of 1987 establishes the federal responsibility for disposal of spent nuclear fuel and high-level waste.

Waste Isolation Pilot Plant Land Withdrawal Act of 1992, as amended, withdraws land from the public domain for operation of the facility; defines operational limitations and the role of the EPA and the US Mine Safety and Health Administration; exempts transuranic mixed waste destined for disposal at the facility from treatment requirements and land disposal prohibitions under the Solid Waste Disposal Act. The Act provides for a continuing EPA oversight role, including recertification that the facility meets EPA standards.

Energy Policy Act of 1992 mandated site-specific public health and safety standards and site-specific licensing requirements for the proposed repository at Yucca Mountain, Nevada. Among other things, it also authorized the DOE to reimburse certain 'active' uranium and thorium milling owners for a portion of their remedial action costs.

Energy Policy Act of 2005 sets forth an energy and development program and includes specific provisions addressing, among other things, disposal of greater-than-class C low-level waste (including certain sealed sources), naturally occurring radioactive materials, and accelerator-produced waste.

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Title 10 (for NRC and DOE) and Title 40 (for EPA) of the US Code of Federal Regulations (CFR) contain the general requirements for the three federal agencies responsible for regulating radioactive waste. US government regulations are developed through an open process, including the opportunity for public comment. New regulations are published in the Federal Register in proposed and final forms.

The separation between the EPA standard-setting function and the NRC's implementing function reflects a nearly 40-year-old congressional policy of centralizing environmental standard-setting in a single agency. When the EPA was established, it was given environmental authorities previously scattered among several older agencies, including the NRC predecessor, the Atomic Energy Commission (AEC). There are advantages to having an agency both set and implement standards, and the NRC does so in many subject areas, especially in reactor design and operation. Nonetheless, there are also advantages to having environmental standards set on a national basis by a single agency whose jurisdiction is wide enough to permit the agency to rank risks from many sources, including nuclear.

18.4 Regulatory oversight: federal and state agencies

18.4.1 Federal agencies

US Nuclear Regulatory Commission

The NRC is an independent regulatory agency created from the former AEC by Congress under the Energy Reorganization Act of 1974 to ensure protection of the public health and safety and the environment, and to promote the common defense and security in the civilian use of byproduct, source, and special nuclear materials. The NRC is authorized to regulate private sector and certain government nuclear facilities, regulating the possession and use of nuclear materials as well as the siting, construction, and operation of nuclear facilities. It performs its mission by issuing regulations, licensing commercial nuclear reactor construction and operation, licensing the possession of and use of nuclear materials and wastes, safeguarding nuclear materials and facilities from theft and radiological sabotage, inspecting nuclear facilities, and enforcing regulations. The NRC regulates commercial nuclear fuel cycle materials and facilities as well as commercial sealed sources, including disused sealed sources.

The NRC regulates:

- commercial nuclear power, nonpower research, test, and training reactors
- fuel cycle facilities and medical, academic, and industrial uses of nuclear materials

- licensing of nuclear waste management facilities (including storage and disposal of SNF and HLW) as well as independent SNF management facilities
- certain DOE activities and facilities over which Congress has provided NRC licensing and related regulatory authority.

The NRC also regulates manufacture, production, transfer or delivery, receiving, acquisition, ownership, possession, and use of commercial radioactive materials, including associated RAW. The key elements of the NRC regulatory program are described in detail at http://www.nrc.gov. In addition, the Department of Transportation has certain regulatory authority over the transport of SNF and HLW. Specifically, the NRC regulates management and disposal of LLW and HLW, as well as decontaminating and decommissioning of facilities and sites. The NRC is also responsible for establishing the technical basis for regulations, and provides the information and technical basis for developing acceptance criteria for licensing reviews.

An important aspect of the NRC regulatory program is inspection and enforcement. The NRC has four regional offices that inspect licensed facilities in their regions, including nuclear waste facilities. Specific information on NRC Regional Offices can be accessed at http://www.nrc.gov/about-nrc/organization.html. The NRC Office of Federal and State Materials and Environmental Management Programs communicates with state, local, and tribal governments, and oversees the Agreement State Program.

US Environmental Protection Agency

The EPA establishes generally applicable environmental standards to protect the environment from hazardous materials and certain radioactive materials. It has authority to establish standards for remediating active and inactive uranium mill tailing sites, environmental standards for the uranium fuel cycle, and environmental radiation protection standards for management and disposal of SNF, HLW, and TRU waste. The EPA promulgates standards for and certifies compliance at the WIPP in New Mexico for disposal of defense-generated TRU waste. EPA standards, under the Clean Air Act (EPA, 1990), limit airborne emissions of radionuclides from DOE sites. The EPA's radioactive waste regulatory functions are described in more detail below.

Other EPA radiation-related authorities

The EPA has regulatory responsibilities for a variety of other man-made and naturally occurring radioactive wastes:

- developing general radiation protection guidance to the federal government
- limiting airborne emissions of radionuclides
- setting drinking water regulations, under the Safe Drinking Water Act (as amended), including standards for radionuclides in community water systems
- coordinating with state radiation protection agencies to protect the environment, workers, and the public from naturally occurring radioactive materials exposed or concentrated by mining or processing
- coordinating with the DOE, NRC, and states on orphaned sources, recycled materials, and controlling imports and exports to prevent radioactively contaminated scrap from entering the United States. The US Coast Guard and the US Department of Homeland Security Customs and Border Protection have the lead in detecting and taking steps to prevent the illegal entry of such materials. They have the authority to take enforcement actions and, depending on the circumstances, may seize or have a shipment returned to the point of origination.

Waste Isolation Pilot Plant oversight

The EPA issues radiation standards and certifies compliance of the WIPP disposal facility. The WIPP Land Withdrawal Act (LWA), as amended, required the EPA to issue final regulations for disposal of SNF, HLW, and TRU waste. It also gave the EPA authority to develop criteria implementing final WIPP radioactive waste disposal standards. The EPA must also determine every five years whether the WIPP facility is in compliance with applicable standards. The WIPP LWA also requires the EPA to determine whether WIPP complies with other federal environmental and public health and safety regulations, such as the Clean Air Act and the Solid Waste Disposal Act. The EPA conducts inspections of both waste generators and WIPP operations. Separate inspections may be conducted for waste characterization activities, quality assurance, or WIPP site activities (procedural or technical).

The EPA also has regulatory authority for radioactive waste disposal standards for SNF, HLW, and TRU radioactive waste; final individual protection standards; final groundwater protection standards; and contamination in offsite underground sources of drinking water.

Mixed waste regulation

A dual regulatory framework exists for mixed waste, which is waste that the EPA considers to be hazardous and radioactive. The EPA or authorized states regulate the hazardous waste component and the NRC, NRC

Agreement States, or DOE regulate the radioactive component. The NRC and DOE regulate mixed waste radiation hazards using Atomic Energy Act of 1954 (AEA) authority. The EPA regulates mixed waste chemical hazards under its Resource Conservation and Recovery Act (RCRA) authority. The NRC is authorized by the AEA to issue licenses to commercial users of radioactive materials.

The EPA issued regulations in 2001 that apply to:

- storage at the generator site or another site operating under the same license
- treatment in a tank or container at the generator site or another site operating under the same license
- transportation to a licensed treatment facility or LLW disposal facility
- disposal at a licensed LLW disposal facility, as long as the waste meets RCRA treatment standards for hazardous constituents.

The EPA has also established National Emission Standards for Hazardous Air Pollutants (NESHAPs) under the Clean Air Act for airborne radionuclide emissions from a variety of industrial sources. Various subparts apply to underground uranium mines, inactive uranium mill tailings piles, and active uranium mill tailings piles, respectively.

US Department of Energy

The DOE has responsibility for, among other matters, nuclear energy, nuclear weapons programs, nuclear and radiological weapons nonproliferation, radioactive waste management, and new nuclear-related activities for environmental remediation of contaminated sites and surplus facilities. It has regulatory authority over its facilities and nuclear activities, and those operated or conducted on its behalf, except where the NRC is specifically authorized by statute to regulate certain DOE facilities and activities.

Specifically, the DOE is responsible for regulating its SNF and RAW management activities pursuant to the AEA, except in cases where Congress has specifically provided the NRC with licensing and related regulatory authority over DOE activities or facilities. Radiation and environmental protection are ensured by a rigorous framework of federal regulations, DOE Orders and Directives, and external recommendations by the Defense Nuclear Facilities Safety Board. The DOE regulates facility operations and radiation protection through standards and requirements established in DOE Orders and Directives.

The DOE implements applicable radiation protection standards considering and adopting, as appropriate, recommendations of authoritative organizations such as the National Council on Radiation Protection and Measurements and the International Commission on Radiological

Protection. It is also DOE policy to adopt and implement standards generally consistent with those of the NRC.

Nuclear Waste Technical Review Board

Congress created the US Nuclear Waste Technical Review Board (NWTRB) in the 1987 amendments to the Nuclear Waste Policy Act (NWPA). It advises both Congress and the Secretary of Energy on technical issues related to DOE implementation of the NWPA. The Board evaluates the technical validity of all activities undertaken by the Secretary of Energy related to DOE's obligation to manage and develop an approach to dispose of SNF and HLW. The NWTRB is a unique federal agency and is completely independent, nonpartisan, and nonpolitical. Its 11 members are appointed by the President from a list of nominees submitted by the National Academy of Sciences, which makes its nominations based solely on the expertise of the individual in relevant scientific and engineering disciplines. The independent technical peer review offered by the NWTRB contributes to the acceptance by the public and scientific communities for different approaches to managing nuclear waste.

18.4.2 State authorities

State regulatory authorities

Provisions of law allow federal agencies to delegate or relinquish certain regulatory responsibilities to the states having radioactive materials or nuclear facilities. NPPs are regulated by federal authorities. Regional arrangements allow closer coordination, such as using radioisotopes for medical uses. These arrangements are not necessarily mandatory; however, where the state can demonstrate adequate competencies, the appropriate federal agency can transfer regulatory authority.

EPA authorized states

The EPA delegates authorities to states in two areas of RAW management. NESHAPs regulations are based on the requirements of the Clean Air Act, and the authority for delegating compliance responsibility to the individual states is described by law. A state must have emission limits at least as stringent as the federal EPA national standards, although most states have not asked for delegation responsibility of radionuclide NESHAPs. The EPA has a similar process for delegating RCRA hazardous waste requirements to states. The state must have a program at least as stringent as the federal program, and the application for authorization must address specific areas of compatibility. For example, the State of New Mexico is authorized by the

EPA to carry out the base RCRA and mixed waste programs in lieu of equivalent federal programs. The New Mexico Environment Department reviews permit applications for treatment, storage, and disposal facilities for hazardous waste under Subtitle C of RCRA. The WIPP Hazardous Waste Facility Permit is renewed every ten years.

States authorized by the EPA play a significant role in regulation and independent oversight of DOE facilities. Most of the DOE's cleanup is performed under the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) through Federal Facility Agreements and under RCRA through various consent and compliance orders. These enforceable regulatory agreements and orders with federal and state agencies establish the scope of work to be performed at a given site and the dates by which specific cleanup milestones must be achieved. Failure to comply with these agreements and orders is subject to fines and penalties.

NRC agreement states

The AEA, as amended, provides a statutory basis for the NRC to relinquish to individual states portions of its authority to license and regulate byproduct materials (radioisotopes), source materials (uranium and thorium), and certain quantities of special nuclear materials. Of the 50 states, 37 have entered into agreements with the NRC to assume this responsibility.

The role of the Agreement States is to regulate most types of radioactive material in accordance with the compatibility requirements of the AEA. These types of radioactive materials include source material (uranium and thorium), reactor fission byproducts, and byproduct materials as defined in Section 11e of the AEA, and quantities of special nuclear materials not sufficient to form a critical mass. The NRC, under its own internal practices, periodically reviews the performance of each Agreement State to ensure compatibility with its regulatory standards.

Agreement States issue radioactive material licenses, promulgate regulations, and enforce those regulations under the authority of each individual state's laws. The Agreement States conduct their licensing and enforcement actions under direction of the governors in a manner compatible with the licensing and enforcement programs of the NRC.

18.5 Waste classification, characteristics, and inventory

18.5.1 Spent fuel and RAW classification

The US classification system has two separate subsystems: one applies to commercial waste, and NRC regulations define it; the other applies to DOE

SNF and waste. The two systems are used for different purposes and different situations so conflicts do not occur. If ownership of radioactive waste is transferred from the DOE to a commercial entity licensed by the NRC, the waste is then subject to NRC regulation (and classification).

Spent fuel

The United States defines 'SNF' as fuel that has been withdrawn from a nuclear reactor following irradiation, the constituent elements of which have not been separated by reprocessing. US law generally uses the term 'SNF' rather than 'spent fuel,' and the DOE has begun using the term 'used fuel' to acknowledge that in the future, the material may have residual value through recycling. For the purposes of this chapter, used fuel is referred to as SNF in accordance with the conventional terminology unless otherwise noted.

Radioactive waste

Radioactive wastes in the United States have many designations depending on their hazards and the circumstances and processes that created them. The NRC regulates most, but not all, sources of radioactivity, including LLW and HLW disposal, and residues from the milling of uranium and thorium. Uranium mill tailings, the final byproduct of uranium ore extraction, are considered radioactive wastes. Radioactivity can range from just above background to very high levels, such as parts from inside the reactor vessel in a NPP. The everyday waste products generated in medical laboratories and hospitals, contaminated by medical radioisotopes, is also designated as RAW.

Tables 18.2 and 18.3 identify the types of commercial and DOE radioactive wastes. NRC regulations classify LLW in the commercial sector as Class A, Class B, and Class C. Radioactive waste owned or generated by the DOE is classified as HLW, TRU waste, or LLW. In addition, the DOE manages large quantities of uranium mill tailings and residual radioactive material. This residual radioactive material, which resulted from the Manhattan Project, is managed under the Uranium Mill Tailings and Radiation Control Act (UMTRCA) Title I. Waste may also contain hazardous waste constituents. Waste with both radioactive and hazardous constituents in the United States is called 'mixed' waste (mixed LLW or mixed TRU waste). Generally, the source of HLW is reprocessed SNF. TRU waste consists of items such as protective clothing, tools, glassware, equipment, soils, and sludge contaminated with man-made radioisotopes beyond or 'heavier' than uranium in the periodic table of the elements.

Table 18.2 US commercial RAW classification

Waste class	Description
HLW	The highly radioactive material resulting from reprocessing of spent fuel, including liquid waste produced directly in reprocessing and any solid material derived from such liquid waste containing fission products in sufficient concentrations and other highly radioactive material that the NRC, consistent with existing law, determines by rule requires permanent isolation.[a]
Class A LLW	Class A waste is determined by characteristics listed in 10 CFR 55(a)(2)(i) and physical form requirements in 10 CFR 61.56(a). (The US does not have a minimum threshold for Class A waste.)
Class B LLW	Waste that must meet more rigorous requirements on waste form than class A waste to ensure stability.
Class C LLW	Waste that not only must meet more rigorous requirements on waste form than Class B waste to ensure stability, but also requires additional measures at the disposal facility to protect against inadvertent intrusion.
GTCC LLW	LLW not generally acceptable for near-surface disposal.
AEA Section 11e. (2) byproduct material	Tailings or wastes produced by the extraction or concentration of uranium or thorium from any ore processed primarily for its source material content, including discrete surface wastes resulting from uranium solution extraction processes. Underground ore bodies depleted by such solution extraction operations do not constitute 'byproduct material' within this definition.[b]

[a]From the Nuclear Waste Policy Act of 1982, as amended.
[b]Title 10 CFR Part 40, *Domestic Licensing of Source Material* (Section 40.4).

Characteristics

SNF results from the once-through fuel cycle (i.e., no further processing conducted). It contains greater than 99% of the radioactivity and has unique characteristics compared to wastes from fossil plants. Because only about 5% of the energy value has been consumed in the reactor, it can also represent a future energy resource. The energy release from nuclear fission per ton of fuel is about a million times greater than the energy release from the burning of fossil fuels. The waste volume generated is about a million times less. The quantity of SNF is small per unit of energy produced. The small quantity (~20 tons per reactor per year) makes multiple waste management options economically feasible: multiple direct disposal options and multiple options to process the SNF chemically for recovery of selected materials for recycle and/or conversion into different waste forms.

Table 18.3 DOE RAW classification

Waste class	Description
HLW	High-level waste is the highly radioactive waste material resulting from the reprocessing of spent nuclear fuel, including liquid waste produced directly in reprocessing and any solid material derived from such liquid waste containing fission products in sufficient concentrations; and other highly radioactive material determined, consistent with existing law, to require permanent isolation.[a]
TRU	Radioactive waste containing more than 3,700 becquerels (100 nanocuries) of alpha-emitting transuranic isotopes per gram of waste, with half-lives greater than 20 years, except for: (1) HLW, (2) waste the Secretary of Energy has determined, with the concurrence of the Administrator of EPA, does not need the degree of isolation required by the 40 CFR Part 191 disposal regulations; or (3) waste the NRC has approved for disposal on a case-by-case basis in accordance with 10 CFR Part 61.
LLW	Radioactive waste not HLW, spent fuel, TRU waste, byproduct material (as defined in section 11(e).2 of the Atomic Energy Act of 1954, as amended), or naturally occurring radioactive material.
AEA Section 11e.(2) byproduct material	The tailings or wastes produced by the extraction or concentration of uranium or thorium from any ore processed primarily for its source material content.

[a]From the Nuclear Waste Policy Act of 1982, as amended.

Reactors discharge SNF that contains fissile materials (fuel) and fission products (waste). The radioactivity and decay heat of SNF decreases rapidly with time; thus, to reduce handling risks and costs, SNF is stored before transport, disposal, or recycling. SNF storage is a required step in all open and closed fuel cycles. This is a consequence of the nuclear characteristics of SNF. The radioactivity decreases rapidly with time, resulting in radioactive decay heat and gamma radiation decreasing rapidly with time. There are large safety and economic incentives to allow the radioactivity of SNF to decrease before transport, processing, or disposal.

Upon reactor shutdown, SNF is intensely radioactive and generates large quantities of decay heat – equal to about 6% of the power output of the reactor. However, the radioactive decay heat decreases very rapidly reaching 0.5% in one week. The refueling strategy in light water reactors (LWRs) is to transfer the SNF from the reactor core to the SNF storage pool where

the water provides cooling and radiation shielding. After about ten years, the radioactivity will decrease by another factor of 100.

If SNF is to be disposed of in a repository, it will likely be stored for approximately 40–60 years prior to disposal. Peak temperatures in a geological repository are limited to ensure long-term repository performance. If the temperatures are too high, the performance of the waste form, waste package, and geology may be impaired. Peak repository temperatures would be controlled by limiting the allowable decay heat per waste package. If the SNF is stored for several decades, several advantages would result: the decay heat per ton of SNF decreases; more SNF can be placed in each waste package; the waste packages can be spaced closer to each other underground; the size (footprint) of the repository is reduced; and the cost of the repository is reduced. Like SNF, the HLW will be cooled for 40–60 years before ultimate disposal to reduce the decay heat.

18.5.2 Spent fuel and high-level waste

Spent fuel storage

The United States produces SNF in commercial NPPs and research reactors. Currently, 104 licensed nuclear power reactors provide about 20% of US electricity. Information on US nuclear power reactors is provided in the Convention on Nuclear Safety US National Report (IAEA, 2012).

All operating nuclear power reactors are storing SNF in NRC-licensed, onsite SNF pools, and over half are storing SNF in NRC-licensed independent spent fuel storage installations (ISFSIs) located onsite. Given the circumstances regarding reconsideration of the US strategy for underground geologic disposal of SNF and HLW and the work performed by the Blue Ribbon Commission (BRC) on America's Nuclear Future (see Section 18.6), the current US approach to SNF management will continue. SNF will remain in onsite storage at the NPP where it was generated in spent fuel pools or at ISFSIs until a national long-term strategy is decided.

Most NPPs that have been decommissioned or are undergoing decommissioning also have SNF stored onsite pending disposal. Most permanently shut-down commercial nuclear power reactors currently have, or are planning to have, their SNF stored at onsite ISFSIs. NRC amended its regulations in 1990 to allow licensees to store SNF in NRC-certified dry storage casks at licensed power reactor sites. Dry storage systems were developed as the preferred alternative (versus new pool construction). Most SNF is loaded in canisters with inert gas and welded closed. The canisters are then placed in storage casks or vaults/bunkers. Some cask designs can be used for both storage and transportation.

There are two primary canister-based, dry-cask storage systems for SNF in the United States (NRC, 2012a). One design involves placing canisters

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vertically or horizontally in a concrete vault used for radiation shielding and protection of the canister. The other design places canisters vertically on a concrete pad and uses both metal and concrete storage overpacks for radiation shielding and canister protection (NRC, 2012b).

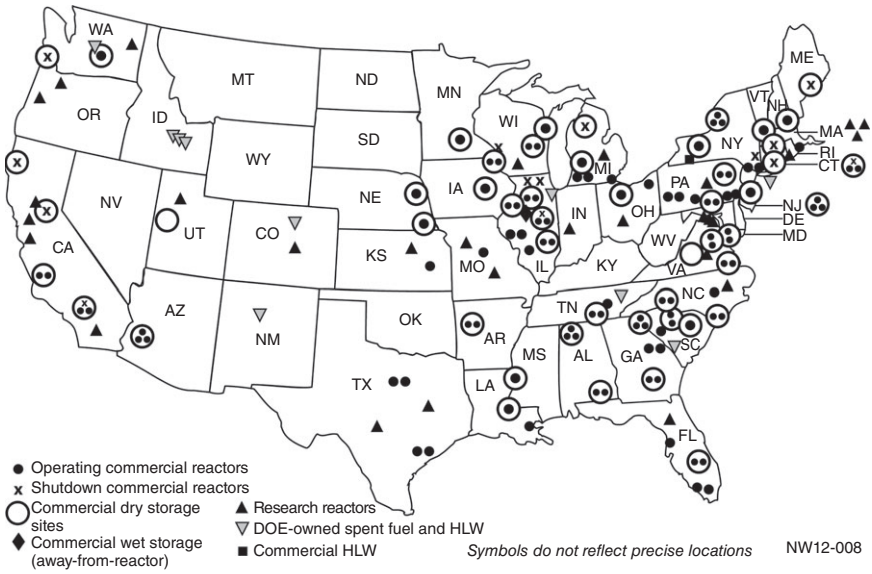
Table 18.4 summarizes the types and numbers of US SNF storage facilities. Complete lists of these facilities of SNF storage facilities are provided in the annex of the United States Fourth National Report for the Joint Convention on the Safety of Spent Fuel Management and on the Safety of Radioactive Waste Management (the complete document can be found at: http://www.em.doe.gov/pdfs/4th_US%20_Nat%20_Report%20%2009-21-11.pdf). Fig. 18.2 shows the location of independent SNF storage installations and other SNF storage facilities.

Recently, the NRC has renewed the licenses for several ISFSIs for a 40-year term, extending the total storage duration authorized by NRC for

Table 18.4 Spent fuel storage facilities

Function	Number of facilities[a]	Inventory (as of 2010)[b]	Units[c]
Government			
Wet storage	8	34	MTHM
Dry storage[d]	7	2,420	MTHM
University research facilities			
Wet storage	21	1,042	kg U
Dry storage	0	0	kg U
Other research and nuclear fuel cycle facilities			
Wet storage	3	36	kg U
Dry storage	1	102	kg U
Onsite storage at nuclear power plants[e]			
Wet storage	68	49,067	MTHM
Dry Storage	52	15,357	MTHM

[a]In some instances, multiple facilities at a given installation are counted as a single facility (e.g., in the case of shared storage pools or independent spent fuel storage installations).
[b]Additional inventory tables can be found in the United States Fourth National Report for the Joint Convention on the Safety of Spent Fuel Management and on the Safety of Radioactive Waste Management at http://www.em.doe.gov/pdfs/4th_US%20_Nat%20_Report%20%2009-21-11.pdf.
[c]MTHM = metric tons of heavy metal.
[d]Includes NRC-licensed facilities at the DOE Idaho Site and Fort St. Vrain in Colorado.
[e]Includes GE Morris and Utah Private Fuel Storage, which are not located at a nuclear power source.



18.2 Location of US spent fuel and HLW storage installations.

60 years. The NRC determined that the licensees' aging management plans along with their surveillance activities were sufficient to ensure that the SNF can be safely stored and retrieved at the end of the 60-year storage period (NUREG, 2011).

Spent fuel disposal

The NWPA of 1982 established the federal responsibility for the disposal of SNF and HLW. The NWPA assigned responsibilities for the disposal of SNF and HLW to three federal agencies:

1. DOE for developing permanent disposal capability for SNF and HLW
2. EPA for developing generally applicable environmental protection standards
3. NRC for developing regulations to implement EPA standards; deciding whether to license construction, operation, decommissioning, and closure of the repositories; and certifying packages used to transport SNF and HLW to the licensed repositories.

The NWPA, as amended in 1987 (Nuclear Waste Policy Amendments Act), directed the DOE to characterize a site at Yucca Mountain, Nevada, for its potential use as a deep geological repository. The geology at Yucca Mountain is a welded volcanic tuff and the climate is arid desert. (Other sites in salt and basalt had previously been under consideration.) However, in 2009,

the Obama Administration decided that Yucca Mountain was no longer an option to be considered (see Section 18.6).

18.5.3 Radioactive waste storage and treatment

Radioactive wastes are treated primarily to produce a structurally stable, final waste form and minimize the release of radioactive and hazardous components. The United States does not commonly make a distinction between the terms 'treatment' and 'conditioning.' Conditioning is defined in the international community as an operation producing a waste form suitable for handling, such as conversion of a liquid to a solid, enclosure of the waste in containers, or overpacking. Treatment is defined as those operations intended to improve the safety and/or economy by changing the characteristics of the waste through volume reduction, removal of radionuclides, and change in composition. US terminology covering both conditioning and treatment is generally referred to as treatment or processing. Treatment is used in this broader context in this chapter.

18.5.4 High-level waste

HLW from commercial reprocessing activities has been vitrified and is stored at the former reprocessing plant in West Valley, New York. Defense HLW is stored, managed, and treated at three DOE sites: Savannah River Site (SRS) in South Carolina, Hanford Site in Washington, and Idaho National Laboratory (INL) in Idaho.

18.5.5 Low- and intermediate-level waste

Low-level waste

Commercial and government facilities exist for LLW processing, including treatment, conditioning, and disposal. Generators prepare LLW for shipment to licensed disposal facilities. Commercial LLW disposal facilities are designed, constructed, and operated under licenses issued by either the NRC or an Agreement State, based on NRC health and safety regulations governing waste disposal quantities, forms, and activity levels. The DOE operates disposal facilities for LLW that it owns or generates and uses commercial LLW disposal sites in certain circumstances.

LLW is disposed of in near-surface facilities, i.e. a land disposal facility in which radioactive waste is disposed of in or within the upper 30 m of the Earth's surface. Currently, commercial generators of Class B and C wastes in 36 states do not have access to a disposal site for these wastes, which are being stored pending a disposal pathway.

Greater-than-class C LLW

Greater-than-class C (GTCC) LLW waste is a form of LLW containing long- and short-lived radionuclides with properties requiring a more robust disposal strategy than for other classes of LLW. In the context of this chapter, 'more robust' means a greater degree of isolation, durability, and performance than is associated with near-surface disposal for other classes of LLW. This could include intermediate-level waste, as defined by some nations. The authority to possess this type of radioactive material is included in NRC or Agreement State licenses.

GTCC LLW may generally be grouped into the following three types: sealed sources, activated metals, and other waste. Other GTCC LLW includes contaminated equipment, trash, and scrap metal from miscellaneous industrial activities, such as manufacturing of sealed sources and laboratory research. Most GTCC LLW is activated metal, generated by decommissioning NPPs, and disused sealed sources. Although the US inventory of GTCC LLW is modest, the construction of new commercial reactors and other proposed actions could generate additional quantities of GTCC LLW. GTCC LLW is stored until an adequate method of disposal is established by the DOE.

Low-level waste (near surface) disposal

There are currently three active, licensed commercial LLW disposal sites. A fourth licensed site currently has facilities under construction:

1. EnergySolutions/Chem-Nuclear (Barnwell, South Carolina). As of July 2008, access is limited to LLW generators within three states composing the Atlantic Compact (South Carolina, Connecticut, and New Jersey). Barnwell disposes of Class A, B, and C LLW up to 0.37 TBq (10 Ci) (which precludes many higher activity sealed sources).
2. US Ecology (on the Hanford Site). Restricted access to only the Northwest and Rocky Mountain Compacts. The member states of the Northwest Compact are Alaska, Hawaii, Idaho, Montana, Oregon, Utah, Washington, and Wyoming. The Rocky Mountain Compact members are Colorado, Nevada, and New Mexico. US Ecology disposes of Class A, B, and C LLW. The US Ecology site can also accept radium and other naturally occurring radioactive materials and accelerator-produced radioactive waste without compact restrictions.
3. EnergySolutions (Clive, Utah). Accepts Class A LLW and mixed LLW for LLW generators without access to other compact facilities.
4. Waste Control Specialists (WCS) (near Andrews, Texas). Provides Class A, B, and C LLW disposal to generators within the Texas Compact (Texas and Vermont). The site is privately owned and regulated by the

State of Texas. Construction began in January 2011, and operations began in April 2012. The Texas Compact has a process in place to accept (import) a limited volume of waste from out-of-Compact states. In addition, WCS constructed a separate facility for disposal of Federal (limited primarily to DOE) mixed LLW and LLW.

Commercial LLW sites now closed are Beatty, Nevada (closed 1993); Maxey Flats, Kentucky (closed 1977); Sheffield, Illinois (closed 1978); and West Valley, New York (closed 1975).

18.5.6 Transuranic waste

TRU waste is managed by the DOE. Defense TRU waste is disposed of in the WIPP geological repository and consists of two types. Remote-handled (RH) TRU waste emits more radiation than contact-handled (CH) TRU waste and must be both handled and transported in shielded casks. Section 18.7.7 provides more details on TRU waste and the WIPP facility.

18.6 Blue Ribbon Commission

In 2009, the Obama Administration announced that it had determined that developing a repository at Yucca Mountain, Nevada, is not a workable option and that the United States needs a different solution for nuclear waste disposal. The Secretary of Energy established the BRC on America's Nuclear Future in January 2010 to evaluate alternative approaches for managing SNF (referred to as 'used nuclear fuel' in BRC documents) and HLW from commercial and defense activities.

The BRC conducted a comprehensive review of policies for managing the back end of the nuclear fuel cycle. It has provided recommendations for 'developing a safe long-term solution to managing the Nation's used nuclear fuel and nuclear waste.' An interim draft report was issued in July 2011, and a final report was submitted to the Secretary of Energy in January 2012 (BRC, 2012).

The report contains eight recommendations for legislative and administrative action to develop a 'new' strategy to manage nuclear waste:

1. A new, consent-based approach to siting future nuclear waste management facilities.
2. A new organization dedicated solely to implementing the waste management program and empowered with the authority and resources to succeed.
3. Access to the funds nuclear utility ratepayers are providing for the purpose of nuclear waste management.
4. Prompt efforts to develop one or more geological disposal facilities.

5. Prompt efforts to develop one or more consolidated storage facilities.
6. Prompt efforts to prepare for the eventual large-scale transport of SNF and HLW to consolidated storage and disposal facilities when such facilities become available.
7. Support for continued US innovation in nuclear energy technology and for workforce development.
8. Active US leadership in international efforts to address safety, waste management, nonproliferation, and security concerns.

The near-term direction advocated by the BRC aligns with ongoing DOE programming and planning. Current programs will identify alternatives and conduct scientific research and technology development to enable long-term storage, transportation, and geological disposal of SNF and all radioactive wastes generated by existing and future nuclear fuel cycles. The BRC report has informed the Administration's work with Congress to define a responsible and achievable path forward to manage used nuclear fuel and nuclear waste in the United States.

In January 2013, the Secretary of Energy issued the Administration's *Strategy for the Management and Disposal of Used Nuclear Fuel and High-Level Radioactive Waste*. The strategy is a 'framework for moving toward a sustainable program to develop an integrated system capable of transporting, storing, and disposing of used nuclear fuel and high-level radioactive waste from civilian nuclear power generation, defense, national security and other activities' (DOE, 2013). It addresses several issues: it serves as an Administration policy statement for handling the disposition of nuclear waste; it presents the response to the BRC report; and it represents an initial basis for discussions among the Administration, Congress, and other stakeholders on the path forward for nuclear waste disposal.

The strategy includes a phased, adaptive, and consent-based approach to siting and implementing a comprehensive management and disposal system. With the appropriate authorizations from Congress, the Administration plans to implement a program over the next ten years that:

- sites, designs, licenses, constructs, and begins operations of a pilot interim storage facility by 2021 with an initial focus on accepting used nuclear fuel from shut-down reactor sites;
- advances toward the siting and licensing of a larger interim storage facility to be available by 2025 that will have sufficient capacity to provide flexibility in the waste management system and allow for acceptance of enough used nuclear fuel to reduce expected government liabilities;
- makes demonstrable progress on the siting and characterization of geologic repository sites to facilitate the availability of a geologic repository by 2048.

The Administration, through the DOE, is undertaking activities within existing Congressional authorization to plan for the eventual transportation, storage, and disposal of used nuclear fuel. Activities range from examining waste management system design concepts, to developing plans for consent-based siting processes, to conducting research and development on the suitability of various geologies for a repository.

18.7 Radioactive waste (RAW) management strategies

18.7.1 Nuclear research and test facilities

SNF from both domestic and foreign research reactors, in addition to limited quantities of commercial SNF, is stored at facilities at the SRS and the INL prior to further disposition. The DOE continues to receive SNF from foreign and domestic research reactors, but plans to complete the program for receipt of foreign research reactor SNF in 2019. No date has been set for completing receipt of SNF from domestic research reactors. The DOE also stores SNF from former defense production reactors. Its current policy and planning includes managing foreign research reactor SNF for 40 years or until ultimate disposition.

18.7.2 Fuel manufacturing

Enrichment and fuel fabrication facilities waste

The product from uranium recovery facilities is processed to enrich the fissile content. Tailings containing depleted uranium (DU) are a byproduct of the enrichment process. Fuel manufacturing facilities fabricate nuclear fuel assemblies for LWRs containing low-enriched uranium. This activity includes receipt, possession, storage, and transfer of special nuclear material. Other licensed activities supporting fuel manufacturing include uranium storage, scrap recovery, waste disposal, and laboratory services. Radioactive waste from these processes, which varies in type and amount, is managed within the classes described in Table 18.2.

Depending on available quantities and long-term and short-term needs, DU could be a resource for a variety of applications and uses, in which case it is considered source material. If DU is not a resource, the NRC categorizes it as Class A LLW. When 10 CFR Part 61 was developed, the disposal of large quantities of DU was not anticipated. However, with the recent licensing of fuel enrichment facilities, which will produce large quantities of DU waste, NRC determined it appropriate to revisit the issue. Therefore, NRC is examining whether the disposal of large quantities of DU from enrichment plants warrants additional, site-specific disposal protections to ensure long-term safety. As an interim measure, the NRC has issued interim

guidance to states that regulate the disposal of large quantities of DU (NRC, 2010).

The DOE and private corporations (e.g., United States Enrichment Corporation) currently possess and store DU. The DOE manages a large stock of DU at two gaseous diffusion enrichment plants, and continues to manage it as source material available for reuse. If a decision is made that this material has no potential use, it can be disposed of in DOE or commercial LLW disposal facilities, provided the waste meets the disposal facility's waste acceptance requirements. Some DOE DU has been disposed of as LLW at the Nevada National Security Site (NNSS), formerly the Nevada Test Site.

18.7.3 Uranium mines and mills facilities

Uranium recovery

Uranium recovery is the extraction or concentration of uranium from any ore processed primarily for its source material content. Similarly, thorium was also extracted or processed in the past. The uranium recovery processes result in wastes that typically contain relatively low concentrations of radioactive materials having long half-lives. The wastes, in both solid and liquid forms, are classified as 11e(2) byproduct material in accordance with AEA definitions (see Table 18.3).

Three types of uranium recovery facilities have operated, are currently operating, or are planned to operate in the future within the United States: conventional mills, heap leach facilities, and *in-situ* recovery facilities. Conventional mills and heap leach facilities extract uranium from ore processed above ground and, consequently, generate large volumes of solid 11e(2) byproduct material. This material is disposed of in licensed near-surface impoundment(s) on the site of the processing facility or in an offsite waste disposal facility licensed to accept 11e(2) byproduct material. *In-situ* recovery facilities differ from the others in that they leach uranium from ore bodies in the subsurface. Consequently, the predominant waste stream for *in-situ* recovery facilities consists of liquid wastes generated during their operation (typically less than 200 megaliters per year). The liquid wastes are disposed of by deep disposal well injection, by evapotranspiration to the atmosphere through land application of partially treated liquid waste, or by evaporation to the atmosphere from man-made lined ponds. The volume of solid waste generated at an *in-situ* recovery facility (including salts from the evaporation process) is relatively small (typically less than 1000 m^3 per year) and is ultimately disposed of offsite at a waste disposal facility licensed to accept 11e.(2) byproduct material.

Prior to the mid-1980s, the sole type of uranium recovery facility in the United States was the conventional mill. Many of those previously operating facilities were reclaimed or are in the process of remediating

(decommissioning) waste resulting from extracting uranium. Because of near-surface impoundments, those properties (and heap leach facilities) will be subject to long-term care after closure through government ownership. *In-situ* recovery facilities do not include onsite disposal impoundments and, thus, do not require long-term care after closure.

Uranium mining and milling

The Uranium Mill Tailings and Radiation Control Act (UMTRCA), which amended the AEA, directed the EPA to establish standards for active and inactive uranium and thorium mill sites. The standards for active sites, issued in 1983 as 40 CFR Part 192 (and amended in 1995), establish limits on radon emanations from tailings as well as contamination limits for buildings, soil, and groundwater. A key aspect of UMTRCA is that it required EPA standards to address nonradiological contaminants in a manner consistent with EPA requirements for managing chemically hazardous waste.

The AEA does not identify uranium-mining overburden as radioactive material to be controlled, and neither the NRC nor the DOE regulate the disposition of conventional mining wastes as part of the nuclear fuel cycle. Once uranium mining product is processed or is brought into the milling circuit, including production from *in-situ* recovery operations, the NRC and Agreement States regulate its possession, use, transport, etc.

18.7.4 Spent nuclear fuel

SNF storage

There are several options for long-term storage of SNF. The three major options for LWR SNF are pool storage at the reactor or a centralized site, dry cask storage at the reactor or a centralized site, and storage/disposal in a repository. All can provide long-term, safe SNF storage. Centralized storage has become the preferred option for many countries (e.g., France, Japan, and Sweden) with significant nuclear power programs.

The current fuel cycle in the United States is an open (or once-through) fuel cycle. Nuclear fuel makes a single pass through a reactor, after which the SNF is removed, stored for a period, and then directly disposed of in a geological repository for permanent isolation. Other fuel cycles (partial recycle or closed fuel cycle) are currently under evaluation but no deployment date has been established. The disposal of SNF and HLW has been a technical and institutional challenge for the United States. However, the United States has successfully sited and operated WIPP – a geological repository for the disposal of defense transuranic (plutonium) wastes – for over a decade.

Dry cask storage is currently the preferred option for long-term storage of SNF because the cask has no moving parts (natural circulation

air-cooling for decay heat removal) and requires very little maintenance. As with transport casks, there are economic incentives to storing the fuel in the pool for a decade before transfer to dry cask storage.

The possibility of storage for a century, which is longer than the anticipated operating lifetimes of nuclear reactors, suggests that the United States should move toward centralized SNF storage sites, starting with SNF from decommissioned reactor sites and in support of a long-term SNF management strategy. Ideally, such storage sites would be at repository sites or at sites capable of future expansion to include reprocessing and other back-end facilities should the United States choose a closed fuel cycle. While this proposal is made in the context of a better long-term fuel cycle system, it also addresses two near-term issues: SNF at decommissioned sites and federal liability for SNF storage.

The federal liability for SNF storage is a result of changing federal policies and delays in the repository program. At the time when most US NPPs were built, it was assumed that LWR SNF would be reprocessed. The plants were built with limited SNF storage capacity because of the expectation that SNF would be shipped within a decade to reprocessing plants for recovery and recycle of plutonium.

US government decisions in the 1970s not to allow commercial reprocessing and the resultant national decision to dispose of SNF directly ultimately led to a decision to ship SNF from reactors directly to a geological repository. Under the NWPA, utilities signed contracts with the federal government for disposal of SNF with removal of SNF from reactor sites starting in 1998. As reactor SNF storage pools filled and it became evident that the US government would not meet its contractual obligations to receive SNF, utilities began to construct modular dry-cask storage systems for their SNF to enable continued operation of the reactors.

There is a growing national obligation to utilities to address the inability of the government to remove SNF from nuclear plant sites, according to contracts signed with the DOE. The costs are meant to cover the expenses utilities have incurred to build their own dry cask storage facilities at their sites. By 2020, most of the utilities will have built their own ISFSIs for which the government will have to pay as required by court decisions.

The Private Fuel Storage Company (PFS), a utility consortium designed and licensed as an ISFSI in Utah, is a limited liability company (LLC) formed from eight commercial nuclear utilities that attempted to establish an interim waste storage facility on the Skull Valley Goshute Reservation in Utah. The project proposed to store 40,000 metric tons of irradiated fuel in dry cask containers above ground on concrete pads.

The NRC issued a license to PFS on February 21, 2006, but conditioned construction authorization on the company first arranging for adequate funding. On February 21, 2007, progress in developing the facility was indefinitely delayed by actions of the US Department of the Interior, which

disapproved the lease arrangement between PFS and the Skull Valley Band and denied PFS the use of public lands for an intermodal transfer facility. The 10th Circuit Court of Appeals vacated decisions by the US Department of the Interior that blocked construction of PFS in June 2010. The ruling returned the PFS application for a right-of-way and lease of tribal land to the Department of the Interior for further consideration. The Department of the Interior was still considering the request in December 2012, when PFS submitted a letter to the NRC requesting that the license be terminated to avoid future licensing fees.

18.7.5 Radioisotope production

Waste disposition for commercial medical isotope production

The DOE/National Nuclear Security Administration (NNSA) is working to accelerate commercial production of the medical isotope molybdenum-99 (Mo-99) in the United States without the use of highly enriched uranium (HEU). Mo-99's primary uses include the detection of disease, including heart disease and cancer, and the study of organ structure and function. The isotope's short half-life and excellent binding properties make it uniquely suited for medical procedures. However, its 66-hour half-life prevents it from being stockpiled during periods of shortage. Mo-99 is a crucial radioisotope used in approximately 80% of all nuclear medicine diagnostic procedures and in roughly 50,000 diagnostic and therapeutic nuclear medicine procedures performed every day in the United States.

In cooperation with commercial partners and the US national laboratories, DOE/NNSA is supporting the US private sector in developing independent, non-HEU-based technical pathways to produce Mo-99 in the United States by 2014. The NRC or Agreement State would have to license any new commercial production facility. The expected waste streams from the production of Mo-99 are likely to include radioactive waste for which there is currently no commercial disposal path. The projects are under development, and production has not yet commenced at the time this book was written. However, disposition of specific waste and spent nuclear fuels and targets resulting from Mo-99 production could impact the technical and economic viability of each of the projects. Until a disposal path is identified, producers of this medical isotope would need to provide onsite storage.

18.7.6 Legacy waste from weapons production: tank waste

The DOE and its predecessor agencies generated liquid radioactive waste as a byproduct of processing SNF for the production of nuclear weapons (DOE, 2009). These wastes were stored in large underground tanks at the

Hanford site, SRS, INL, and the West Valley Demonstration Project (WVDP) in New York State. The DOE Office of Environmental Management (EM) is now safely storing 333 million L (88 million gallons) of tank waste in 229 underground tanks at three sites:

1. Hanford: 204 million L (54 million gallons) in 177 tanks
2. SRS: 125 million L (33.1 million gallons) in 49 tanks
3. INL: 3.4 million L (0.9 million gallons) in three tanks.

Tank waste is by far the DOE's most significant environmental, safety, and health challenge, as well as the largest cost element of the cleanup program. Many of these underground tanks, particularly at Hanford, have exceeded their design lives. The DOE expends significant resources and attention to monitoring and maintaining the tanks to ensure they are sound and leak free and that workers can safely perform the necessary tank maintenance and remediation.

The unique and hazardous nature of liquid RAW requires development of innovative technologies for waste retrieval and disposition. These include constructing treatment plants to convert liquid waste into a stable, long-lasting waste form such as glass until it can be safely disposed of in a geological repository. These treatment plants house highly complex chemical and physical treatment processes and must be very robust to operate safely over many years and to protect workers from radiation fields and contamination. Thus, they are expensive to construct and operate and require advanced engineering and technologies.

The strategy for dealing with DOE's tank waste is to:

- minimize the volume of high-activity waste to be solidified through treatment
- store glass canisters onsite until a federal repository is ready for permanent disposal
- solidify the low-activity waste (LAW) fraction and dispose onsite
- develop approaches to manage/treat/dispose of some tank wastes as other than HAW
- continue emptying and closing tanks according to compliance agreements.

Retrieval

The first step in mitigating the risks posed by the tanks is to remove the waste, particularly focusing on the older single-shell tanks (as opposed to an inner and outer double-shell tank with space in between for containing and monitoring any leakage). This was already accomplished at Hanford where nearly 11.3 million L (3 million gallons) of liquids that could be

removed from single-shell tanks physically and cost-effectively were retrieved and moved into double-shell tanks. At other sites, tanks have been emptied to the maximum extent practicable and then backfilled with concrete or grout to stabilize the small amount of contamination remaining. Since 2002, seven 1.1 million L (300,000 gallon) underground storage tanks and four smaller 111,000 L (30,000 gallon) ancillary tanks at the INL have been emptied, cleaned, and filled with concrete. In addition, two 4.9 million L (1.3 million gallon) SRS tanks were closed and grouted in 1997, and an additional two were filled with concrete in 2012.

Tank waste treatment

Once the waste has been retrieved to the maximum extent practicable, the next step is to separate it chemically and physically into two fractions: the higher-volume portion that contains shorter-lived, less radioactive elements (i.e., LAW) and a much smaller fraction that contains longer-lived, radioactive elements (i.e., HAW). The two fractions are then treated separately to convert them to stable, solid forms. The LAW is proposed to be disposed of onsite, and the HAW is proposed to be disposed of offsite in a geological repository.

The Salt Waste Processing Facility (SWPF) and the Waste Treatment and Immobilization Plant (WTP) are being constructed at SRS and Hanford, respectively, to treat and immobilize radioactive tank waste. SRS is completing the design and construction of the SWPF. The SWPF will separate the LAW and HAW fractions, solidifying the former as a grout in the existing Saltstone facility for disposal onsite in large vaults. The HAW fraction will be sent to the Defense Waste Processing Facility (DWPF), which has operated since 1996, where it will be converted to a stable glass form using vitrification. DWPF has vitrified HAW into 3,325 canisters as of December 2011 that are stored onsite in special-purpose facilities awaiting disposal in a geological repository.

To maintain the compliance-driven schedule for closing SRS tanks and to address risk more quickly, SRS began operating two interim tank-waste processing facilities (the Actinide Removal Process and the Modular Caustic Side Solvent Extraction Unit) in advance of SWPF startup to separate out LAW for onsite disposal. The DOE continues to pursue strategies to optimize the capacity of these facilities to complete treatment of the tank waste in a cost-effective manner.

The WTP, now under construction at Hanford, will also separate the LAW and HAW tank fractions. It will then vitrify the two waste fractions, with the LAW disposed of onsite and the HAW disposed of in a geological repository. Operation of the WTP facility is scheduled to begin in 2019.

The remaining INL tank waste will be treated in the Integrated Waste Treatment Unit (IWTU) at the Sodium Bearing Waste Treatment Facility forming a crystalline ceramic (mineral) waste form by fluidized bed steam reforming for ultimate disposal at WIPP. A vitrification plant constructed at West Valley has converted the radioactive tank waste there into 275 canisters of glass.

Tank waste disposal

Until a repository for permanent disposal becomes available, the DOE will store canisters of solidified high-activity tank waste onsite. The stabilized product of LAW treatment at WTP and at Saltstone (facilities for safely stabilizing and disposing of low-level radioactive liquid salt wastes) will be disposed of onsite in stainless steel containers at Hanford and in concrete vaults at SRS, respectively. These wastes contain only 1–10% of the radioactivity present in the tank waste.

Tanks at INL and Hanford contain liquid wastes that are not radioactive wastes generated from the reprocessing of SNF. The DOE plans to pursue alternative but safe, compliant, and more cost-effective disposal paths for these wastes on a case-by-case basis. For example, some may meet the criteria for disposal at the WIPP.

18.7.7 Transuranic waste and the Waste Isolation Pilot Plant

TRU waste is a type of RAW that contains elements with atomic numbers greater than uranium (DOE, 2009). This waste consists primarily of clothing, tools, rags, residues, soil, debris, and other materials contaminated with plutonium; it may also be mixed with hazardous components. There are two categories of TRU waste: CH TRU waste can be handled by workers under very controlled conditions with no shielding for radioactivity other than the container itself, while RH TRU waste must be handled and transported in lead-shielded containers and casks because it emits more penetrating radiation. CH TRU represents 96% of the total volume of TRU waste to be disposed of at WIPP, while RH TRU makes up the remaining 4%.

Before WIPP opened, 28 DOE sites were storing TRU waste in a variety of configurations, primarily below-grade to contain the radioactive elements while also allowing for its eventual retrieval for disposal. After nearly 20 years of testing, scientific research, engineering and design, and regulatory permitting, WIPP began receiving CH TRU waste in 1999. In 2006, WIPP received final authorization to begin accepting RH TRU and the first shipment, from INL, arrived in January 2007.

Located 2,150 feet below ground in a 250 million-year-old salt formation, WIPP is the world's only operating deep geological repository. An estimated 150,000 m^3 of CH TRU and 7,000 m^3 of RH TRU resulting from US Cold War defense activities will ultimately be disposed of there.

Between 2002 and 2008, the DOE de-inventoried all legacy TRU waste at 14 sites, thereby eliminating associated management costs at these sites as well as environment, safety, and health risks. TRU waste was also removed from facilities at the NNSS, Lawrence Livermore National Laboratory, and Argonne National Laboratory (ANL) so they can support other missions.

As of February 2013, WIPP had received 11,112 shipments of TRU waste since it opened in 1999. These years of experience and a streamlined regulatory framework have resulted in more efficient and routine operations with each passing year. The DOE has a clear strategy for building on this past success to meet its TRU risk reduction goals:

- characterize a small quantity of waste in Idaho for shipment to WIPP
- expand use of Central Characterization Project (CCP)
- facilitate shipping sites in certifying waste for acceptance at WIPP
- expand number of sites certified for RH shipping
- deploy shielded containers for shipping RH TRU.

This strategy includes expanding the number of sites certified for RH TRU shipping. To support and enhance this strategy, the DOE continues to develop shielded containers for RH TRU lead-lined drums that allow RH TRU waste to be handled, shipped, and potentially disposed of in a manner similar to CH TRU waste. Currently, RH TRU waste is emplaced in boreholes along the walls of the WIPP repository and CH TRU waste is placed on the floors.

Significant coordination is required for optimal and efficient emplacement of RH TRU and CH TRU waste. The use of shielded containers for placement of selected RH TRU waste on the floors of the repository could increase the efficiency of disposal operations at WIPP. The DOE is actively pursuing the necessary regulatory approvals needed to move forward with shipping and disposing of RH TRU waste in shielded containers at WIPP.

Another TRU waste risk-reduction strategy is the characterization of small-quantity TRU waste sites in Idaho for shipment to WIPP. A Record of Decision (ROD) approved in February 2008 allows the DOE to send waste from small-quantity sites to INL for treatment, characterization, and shipment to WIPP, assuming the waste meets INL waste acceptance criteria. This reduces costs by eliminating the need to construct TRU waste treatment facilities at sites with small quantities of TRU waste. It also results in faster removal of TRU from these sites and a greater economy of scale for the TRU waste facility at INL.

The DOE is also expanding the use of the CCP at large sites. The project employs a modular waste characterization system consisting of full disposal characterization equipment for both CH TRU and RH TRU waste and a mobile loading system used to place drums of TRU waste into shipping containers for transport to WIPP. CCP has proven successful in characterizing waste more cost effectively through use of a standard suite of procedures, quality assurance documents, and equipment.

Another strategy includes the use of TRU waste expert teams to assist generator sites in certification and characterization planning for waste streams that are more difficult to manage, such as those requiring additional documentation, treatment, or packaging. These teams help to ensure all TRU waste is characterized, shipped, and disposed of at WIPP.

The DOE has designed a new cask, TRUPACT-III, for TRU waste packaged in large boxes that cannot be shipped in currently available transportation casks due to their size. The strategy to ship and dispose of large-size containers at WIPP also requires the development, deployment, and regulatory approval of equipment needed to determine the contents of large containers. With this knowledge, the potentially dangerous and costly task of reducing the size of large containers before shipment and disposal at WIPP can be avoided.

18.7.8 Low-level waste and mixed low-level waste

LLW is radioactively contaminated material that is not HLW, SNF, TRU, byproduct material, or naturally occurring radioactive material (DOE, 2009). Under the AEA, the DOE is self-regulating with regard to LLW. Mixed low-level waste (MLLW) is LLW that also contains a hazardous component and is, therefore, subject to a dual regulatory framework, under the AEA, including DOE Order 435.1, Radioactive Waste Management, as well as federal or state hazardous waste requirements promulgated under RCRA (DOE, 1999).

The strategy to deal with LLW and MLLW is:

- continue to utilize a combination of DOE onsite, DOE regional, and commercial disposal facilities
- complete an Environmental Impact Statement (EIS) for commercial GTCC waste and issue ROD for GTCC disposal facility
- reuse/disposition contaminated nickel
- build new onsite CERCLA cells
- continue to pursue treatment alternatives for wastes currently incinerated at the Toxic Substances Control Act Incinerator at the Oak Ridge Reservation in Tennessee
- continue to develop disposition plans for remaining legacy MLLW and LLW, eliminating waste acceptance and/or transportation barriers.

The DOE produced the *Final Waste Management Programmatic Environmental Impact Statement (EIS) for Management, Treatment, Storage, and Disposal of Hazardous Waste* in 1997 (DOE, 1997). The associated complex-wide decisions for treatment and disposal of LLW and MLLW were issued in 2000. These documents described the approach EM would use to eliminate the inventory of legacy LLW and MLLW, the latter in accordance with applicable regulatory agreements. As Table 18.5 illustrates, the DOE has an estimated 1.2 million m^3 of LLW and MLLW.

While treatment and disposal of most LLW and MLLW are now routine, the DOE has inventories of both that lack readily available disposition options. The DOE is focusing on developing pathways for this waste. One category of waste for which a disposal solution has been developed is 'silo material', generated at the Fernald Site in Ohio. This waste was a byproduct of uranium processing, and the radium it contained emitted large amounts of radon. As a result, it was stored in heavily shielded concrete silos. Because of the nature of this material and the regulatory framework surrounding it, it required a specialized license.

The DOE worked closely with a vendor and state regulators in Texas to allow storage of the Fernald silo material at a Texas commercial facility. Removal of the silo material allowed the DOE to close the Fernald site on schedule in 2006 and greatly reduce the environmental risk of continued storage there. The vendor subsequently applied for a disposal license for this type of material and received the requested permit from Texas regula-

Table 18.5 Disposal of low-level waste and mixed low-level waste

Facility	Waste type	Amount of waste
Onsite disposal – INL, SRS, ORR, and LANL	LLW	
Regional disposal – Hanford and NNSS	LLW and MLLW	
Commercial disposal facilities (when cost effective and in the interest of the federal government)	LLW and MLLW	
Legacy and newly generated waste in the DOE Environmental Management program	LLW and MLLW	1.2 million m^3
Environmental restoration cleanup (DOE sites) – Fernald, Hanford, INL, and ORR	LLW and MLLW	6 million m^3
Environmental restoration cleanup (commercial sites)	LLW and MLLW	3 million m^3

INL = Idaho National Laboratory; SRS = Savannah River Site; ORR = Oak Ridge Reservation; LANL = Los Alamos National Laboratory; NNSS = Nevada National Security Site.

tors in 2008. The disposition path for the Fernald silo material is now finalized and approved.

To complete cleanup of the Rocky Flats Plant in Colorado, the DOE supported technology development to decontaminate 1,500 gloveboxes sufficiently to allow equipment to be disposed of as MLLW or LLW. Gloveboxes are sealed chambers in which workers handle plutonium using long rubber gloves that extend through portholes. They range in size and can be as large as a bus. Previous disposition plans called for the gloveboxes to be reduced in size (cut into smaller pieces), packaged, characterized, and certified for disposal at WIPP. This revised approach significantly reduced work exposure to contamination, workplace hazards, and associated costs.

DOE EM has the lead for developing the EIS for the disposal of GTCC low-level radioactive waste and GTCC-like waste. GTCC waste is LLW resulting from US NRC-licensed activities with radionuclides that would be dangerous to humans beyond 500 years. This waste stream comprises materials such as radioactive sources commonly used to sterilize medical products, detect flaws and failures in pipelines and metal welds, and serve other industrial and medical purposes. These materials were generated, owned, or managed by commercial entities rather than the DOE. However, the Low-level Radioactive Waste Policy Amendments Act of 1985 assigned the federal government responsibility for the disposal of certain GTCC radioactive waste resulting from US NRC-licensed activities.

GTCC waste is the highest radiological activity waste with no planned disposition path. The DOE is preparing an EIS to evaluate disposal options for commercial GTCC LLW as well as LLW similar in character to GTCC generated by the DOE. The DOE issued a Notice of Intent to prepare the EIS in July 2007. A draft EIS was issued by the DOE in February 2011, and a final EIS is expected to be released in 2013. By law, before the DOE makes a final decision on the disposal alternative(s) to be implemented, the agency must submit a report to Congress and await Congressional action before making a final disposal decision.

Contaminated nickel from the shutdown of gaseous diffusion plants is a potentially valuable asset. The DOE is evaluating the feasibility of recovering the nickel for potential sale to an end user rather than disposing of it as LLW.

18.8 Site cleanup and closure experience

18.8.1 US experience

For over five decades, the United States generated a large quantity and variety of nuclear wastes. Significant progress has been made in the treatment and disposal of these wastes and the cleanup and closure of nuclear

sites. Much has been accomplished, but work remains to be done before the cleanup mission is complete.

The DOE has over 20 years of experience in site cleanup. DOE EM manages the DOE cleanup program, which has:

- stabilized millions of liters/gallons of radioactive tank waste
- completed 11 waste tank closures, including two in 2012 at the SRS in South Carolina
- operated the DWPF at the SRS since 1996 making 5,850 metric tons of borosilicate glass, which stabilized 1.5×10^6 Tera-Becquerels of radioactivity
- operated and completed waste processing at the West Valley Demonstration Project (WVDP) in New York from 1996 to 2002 making ~500 metric tons of borosilicate glass which stabilized 9×10^5 Tera-Becquerels of radioactivity
- begun construction of three major tank waste processing facilities.

The tank waste processing facilities include the WTP in Washington (2003), SWPF in South Carolina (2005), and the Sodium Bearing Waste Treatment Facility in Idaho (2003). The IWTU at the Idaho facility is expected to begin operations in 2013. See Section 18.7.6 for more detail about these three construction projects.

In addition, the world's first geological repository – WIPP – began operations in 1999, and had received over 11,000 shipments as of February 2013. The first CH TRU waste shipment arrived at WIPP from Los Alamos in 1999, and the first RH waste shipment arrived at WIPP from Idaho in 2007.

The DOE has also treated 240 km^2 of contaminated groundwater and stabilized more than 180 contaminated groundwater plumes. It has extensive experience in deactivation and decommissioning (D&D), including D&D of about 1,500 facilities. For example, it is in the process of decommissioning and demolishing the K-25 facility in Tennessee, a building nearly one mile long used to enrich uranium from 1945 to 1964. It contained nearly 5 million ft^2 of floor space. Demolition of the west wing, which comprises just under half of the entire facility, began in 2008 and finished in 2010.

Another example of a completed D&D activity is the P Reactor in South Carolina (which was entombed in place using concrete grout to fill the rooms below ground level), disassembly basin, and reactor vessel. Cleanup of the Experimental Breeder Reactor-II in Idaho, which operated for about 30 years from the mid-1960s to the mid-1990s, is currently in progress. The systems and structures above the reactor building will be demolished and most of the remaining systems and structures will be grouted in place.

Other D&D projects include the K-Basins project and N Reactor closure in Washington. The K-Basins stored spent fuel; they were demolished in 2009, and remediation of the nearby soil was completed in 2010. N-Reactor

operated from 1963 to 1987; its support facilities have been demolished, and it is being placed into safe interim storage.

The DOE has experience in LLW disposal. At the Hanford site, the Environmental Restoration Disposal Facility began operation in 1996 to dispose of contaminated soils, D&D waste, asbestos, and hazardous waste from onsite cleanup. Waste is disposed in cells approximately 150×150 m in area and about 20 m deep. Another LLW disposal facility at the Oak Ridge Reservation in Tennessee, the Environmental Management Waste Management Facility, has been operating since 2002.

The DOE has closed two former nuclear sites: the Rocky Flats Plant in 2005 and the Fernald Site in 2006. The Rocky Flats Plant was established in 1951 as part of the US nuclear weapons complex to manufacture nuclear weapons components. The site covers about 6,500 acres near the Rocky Mountains northwest of Denver. Most of the land served as a security buffer around an approximately 400-acre industrial area near the center of the site. When production of weapons components ended at Rocky Flats in 1994, its mission changed to cleanup and closure.

Because of operational problems and practices during the plant's history, facilities contained substantial amounts of hazardous materials and contamination. Liquids remained in process piping and in tanks in unknown quantities and chemical configuration, which resulted in a significant environmental cleanup and closure challenge for the DOE.

In October 2005, the DOE and its contractor completed an accelerated ten-year, $6.7 billion cleanup of chemical and radiological contamination left from nearly 50 years of production. The cleanup required the decommissioning, decontamination, demolition, and removal of more than 800 structures, including six processing and fabrication building complexes; removal of more than 500,000 m^3 of LLW; and remediation of more than 360 potentially contaminated environmental sites. The majority of the property at the site was transferred to the US Department of Interior for management by the US Fish and Wildlife Service as the Rocky Flats National Wildlife Refuge in July 2007 (DOE, 2011a).

The Fernald site, formally known as Feed Materials Production Center, was a uranium processing facility that produced high-purity uranium metal products as the first step in the US nuclear weapons production cycle. The site's production mission began in 1951 and continued until 1989, when production operations ceased and Fernald's mission changed to environmental remediation. The comprehensive environmental remediation and ecological restoration of the site was completed in 2006, at a total cost of $4.4 billion.

The 1,050-acre site, now known as the Fernald Preserve, is open to the public as a nature preserve. The ecological restoration has made the Fernald Preserve attractive to a large number of nesting and migrating birds,

including locally rare species. Restoration activities at the site have created one of the largest man-made wetlands, including open water, forests, 360 acres of grassland, and seven miles of trails that provide access to varied habitats (DOE, 2011b).

Significant challenges remain in the DOE cleanup program. The DOE must safely store, retrieve, and treat approximately 340 million L (about 90 million gallons) of liquid radioactive waste stored in 230 underground tanks, remediate approximately 6.5 trillion L of contaminated groundwater, remediate approximately 40 million m^3 of contaminated soil, and D&D over 2,500 facilities.

In addition, the DOE has decommissioned and cleaned up uranium mines and mill tailings. For conventional US uranium mills, waste is primarily the onsite disposal of tailings (residual ore after the uranium was leached). UMTRCA classified the tailings as either residual radioactive material or 11e.(2) byproduct material depending on the status of the facility at the time UMTRCA was passed in 1978. Since passage of UMTRCA, activities at Title I sites have focused largely on decommissioning and cleanup of residual radioactive material by US governmental entities.

UMTRCA Title I required the DOE to complete surface remediation and groundwater cleanup at the listed inactive uranium milling sites at which uranium was processed solely for sale to the US government. Residual radioactive material, including any wind-blown dust, may have been consolidated into a single cell or perhaps relocated to a cell constructed on another site. These cells are now under long-term surveillance by the DOE (or possibly by the state or tribal governments in which the cell is located) and licensed by the NRC. Annual site inspections are performed as part of the long-term surveillance program at 22 Title I disposal sites.

18.8.2 Key elements of the cleanup program and lessons learned

The most important part of the DOE cleanup program is safety, which is integral to every program and project. In addition, DOE EM is implementing DOE Standard 1189 (DOE, 2008), which requires that safety-related documents and reviews be completed in the initial stages of the design process. DOE EM expects that integrating safety analyses up front in project design will avoid costly changes later (DOE, 2009).

Technology development is another key element of the cleanup program. The technology program is designed to provide a best-in-class science and engineering foundation and develop new technologies to reduce technical risk and uncertainty, support cleanup decisions, improve operational efficiency, reduce costs, and accelerate schedules. In addition,

laboratory- and pilot-scale testing is an important part of the technology maturation process.

The EM program has a strong commitment to reducing the technical risk of its programs and projects, and it is implementing two efforts to reduce those risks. This first is to conduct a Technology Readiness Assessment (TRA) to reduce the risks of deployment of a new technology. TRAs provide a snapshot in time of the maturity of technologies and their readiness for inclusion in the project. The results of a TRA assist program and project managers in developing plans to mature the technologies and to make decisions related to technology insertion. Eleven TRAs had been completed by the end of 2012.

The second effort is to conduct an External Technical Review (ETR) as one of several steps to ensure timely resolution of engineering and technology issues. The results of the reviews serve as a basis for developing strategies for reducing identified technical risks, and providing technical information needed to support critical project decisions. Twenty-five ETRs had been completed by the end of 2012.

Adhering to sound project management practices is essential. This includes, but is not limited to, developing comprehensive plans with a clear end-state for the site, defining clear project scopes, identifying and assessing risks, conducting system analyses, conducting peer reviews, establishing firm performance objectives, and anticipating unexpected outcomes.

The cleanup program would not be nearly so successful without the full involvement of its stakeholders, who provide insights and advice on how best to implement and improve it. The program has citizen advisory boards chartered under the Federal Advisory Committee Act at eight cleanup sites. The DOE also supports working groups with the National Governors Association, National Conference of State Legislators, Energy Communities Alliance representing local governments, and State and Tribal Government Working Group. The DOE also works closely with its federal and state regulators to ensure that cleanup is being conducted in accordance with applicable laws, regulations, and compliance agreements, and in ways and according to schedules that protect public health and the environment (DOE, 2010). Continuous and transparent communication with stakeholders is vital.

The DOE's cleanup mission poses unique, technically complex, and costly challenges, which can be achieved only through an exceptional workforce. The program's 40,000 federal and contractor employees have the necessary skills and experience such that it is a world leader in the safe management and disposition of RAW and nuclear materials, as well as the remediation of contaminated facilities, soil, and groundwater (DOE, 2010).

In summary, the United States has extensive experience in cleanup of nuclear waste and facilities resulting from half a century of nuclear

activities. The cleanup program has solved environmental problems that, at one time, seemed unsolvable; it will continue to make progress in solving the complex challenges it still faces (DOE, 2009).

18.9 Yucca Mountain: history and lessons learned

18.9.1 Background

In 1977, the DOE identified Yucca Mountain, Nevada, as a potential repository site for future investigation to host the nation's first deep geological repository for the disposal of SNF and HLW (Fig. 18.3). Other potential sites included bedded salts in Texas and Utah, salt domes in Louisiana and Mississippi, and basalt in the State of Washington. In 1982, Congress passed the NWPA, which established an office within the DOE with the responsibility of providing for the permanent disposal of SNF and HLW, and laid out the process for siting, developing, licensing, and constructing a geologic repository. In 1987, the NWPA was amended and directed the DOE to

18.3 Aerial view of Yucca Mountain, Nevada (Idaho National Laboratory).

investigate only one potential repository site, at Yucca Mountain. The period from 1987 to 2002 was devoted to site characterization of the Yucca Mountain site for a geologic repository, and the following years were dedicated to engineering studies and license application (LA) activities. In February 2002, the Secretary of Energy recommended the site to the President, and the President recommended the site to Congress. In July 2002, Congress granted the authority to the DOE to prepare and submit a LA for constructing a repository at Yucca Mountain. The LA was submitted to the NRC in June 2008, and it was subsequently accepted for review by the NRC.

In early 2009, the Obama Administration determined that a repository at Yucca Mountain was not a workable option and that the project should be terminated. On March 3, 2010, the DOE filed a motion with an NRC Atomic Safety and Licensing Board (ASLB), seeking permission to withdraw the license application for a HLW repository at Yucca Mountain. On June 29, 2010, the ASLB issued an Order denying the DOE's motion to withdraw. This decision was appealed to the NRC. In October 2010, the NRC commenced and continued with the orderly closure of Yucca Mountain LA review activities. In September 2011, the Commission announced that the commissioners were evenly divided on the question of whether the ASLB Order should be overturned but, for budgetary reasons, ordered the ASLB to complete all pending case management matters. The ASLB suspended the licensing proceeding and, as of April 2012, the proceeding remains suspended.

18.9.2 Lessons learned

The NWTRB prepared a comprehensive document in June 2011 that identified lessons learned from Yucca Mountain and other programs (NWTRB, 2011). In its letter to Congress, the Board explained that in the report, it examined 'from a technical perspective the history of the Yucca Mountain program and some other nuclear waste programs and discusses technical information and insights that may be useful for future US high-activity waste management and disposal efforts.'

The Board concluded that the experience gained from the Yucca Mountain program strongly established a technical working base to allow the country to move forward with geological disposal. In addition, the Board acknowledged that international cooperation is an important component in the worldwide challenge for waste management and disposal.

The experience gained from the Yucca Mountain program can be combined and leveraged with existing strengths of the international nuclear community to advance the science of SNF and HLW disposition. A permanent solution is still needed to address the disposal of SNF and HLW in the

Table 18.6 Lessons learned from the US Nuclear Waste Technical Review Board

1. Multiple waste forms exist in the US requiring deep geologic disposal, which could possibly be retrieved in the future, while vitrified HLW would preclude retrieval for further recycling.
2. Nuclear waste can be isolated in a geologic unsaturated zone in an oxidizing environment, and other programs worldwide have identified other potential repository sites in igneous rocks (granite) and sedimentary rocks (e.g., clay and salt).
3. Research in engineered barriers increases the confidence for waste isolation, and delays a dependence on the natural system; with a reduction in the radiotoxicity of waste, the chemistry becomes simplified and enhances the predictive capabilities of long-term performance of a repository.
4. To better assess the performance of a geologic repository and create an efficient design, it is important to quantify the radionuclide source term that could enter the natural system.
5. If the Yucca Mountain project had adopted a 'total integrated systems approach' for performing some of the science and engineering activities, many of the program deficiencies could have been avoided; the transition from a science program to an engineering program requires making critical decisions along the way.
6. Probabilistic performance assessments allow for better risk management and should be used throughout the entire lifetime of the program.

United States, while creating confidence among program stakeholders, protecting the environment, and ensuring the safety and health of the public.

The major lessons learned identified by the Board are listed in Table 18.6.

18.10 Acknowledgement

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18.12 Appendix: acronyms

AEA	Atomic Energy Act
AEC	Atomic Energy Commission
ANL	Argonne National Laboratory
ASLB	Atomic Safety and Licensing Board
BRC	Blue Ribbon Commission
CCP	Central Characterization Project
CERCLA	Comprehensive Environmental Response, Compensation, and Liability Act
CFR	Code of Federal Regulations
CH	contact-handled
D&D	deactivation and decommissioning
DOE	Department of Energy
DU	depleted uranium
DWPF	Defense Waste Processing Facility
EIS	Environmental Impact Statement
EM	Office of Environmental Management
EPA	Environmental Protection Agency
ETR	External Technical Review
GTCC	Greater-than-Class C
HAW	high-activity waste
HEU	highly enriched uranium
HLW	high-level waste
INL	Idaho National Laboratory
ISFSI	independent spent fuel storage installation
IWTU	Integrated Waste Treatment Unit
LA	license application
LAW	low-activity waste
LLC	limited liability company
LLW	low-level waste
LWA	Land Withdrawal Act
LWR	light water reactor
MLLW	mixed low-level waste

NESHAPs	National Emission Standards for Hazardous Air Pollutants
NNSA	National Nuclear Security Administration
NNSS	Nevada National Security Site
NPP	nuclear power plant
NRC	Nuclear Regulatory Commission
NWPA	Nuclear Waste Policy Act
NWTRB	Nuclear Waste Technical Review Board
PFS	Private Fuel Storage Company
RAW	radioactive waste
RCRA	Resource Conservation and Recovery Act
RH	remote-handled
ROD	Record of Decision
SNF	spent nuclear fuel
SRS	Savannah River Site
SWPF	Salt Waste Processing Facility
TRA	Technology Readiness Assessment
TRU	transuranic
UMTRCA	Uranium Mill Tailings and Radiation Control Act
VLLW	very low-level waste
WCS	Waste Control Specialists
WIPP	Waste Isolation Pilot Plant
WTP	Waste Treatment and Immobilization Plant
WVDP	West Valley Demonstration Project

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19

Canada: experience of radioactive waste (RAW) management and contaminated site cleanup

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Abstract: This chapter provides an overview of the policy and regulatory frameworks for radioactive waste in Canada. The chapter then discusses the strategies and long-term management approaches for various classes of radioactive waste generated from selected nuclear sectors, such as nuclear power generation, nuclear research, fuel fabrication, uranium mining, milling, refining and conversion, and radioisotope production and use. Lastly, the chapter provides examples of contaminated site cleanup and decommissioning projects, as well as lessons learned from implementing these projects.

Key words: policy and regulatory frameworks for radioactive waste, contaminated site cleanup, decommissioning, radioactive waste.

19.1 Policies and regulations

19.1.1 Radioactive waste (RAW) policy

The Government of Canada has policies, legislation and responsible organizations that ensure safe management of radioactive waste in Canada. The Government of Canada's *Policy Framework for Radioactive Waste* consists of a set of principles governing the institutional and financial arrangements for management of radioactive waste (Natural Resources Canada, 1996). A key principle within the *Policy Framework* is that waste generators and owners are responsible, in accordance with the principle of 'polluter pays', for the funding, organization, management and operation of long-term waste management facilities and other facilities required for their wastes. The *Policy Framework* recognizes that arrangements may be different for the different categories of radioactive waste in Canada. In the case of nuclear fuel waste, the Government of Canada determined that it would be in the best interests of Canadians to have a national long-term management approach. In 2002, the Government of Canada brought into force the Nuclear Fuel Waste Act (NFWA), which outlines a process for the development and implementation of a long-term management approach for Canada's nuclear

fuel waste and required that an organization, the Nuclear Waste Management Organization (NWMO), be established to carry out the work.

19.1.2 Regulatory framework

In Canada, the management of used nuclear fuel, radioactive waste (RAW) management and uranium mines and mills associated facilities are regulated in a similar fashion. Safety and licensing issues are regulated according to Nuclear Safety and Control Act (NSCA) requirements and associated regulations to ensure that facilities and activities with respect to health, safety, security and the environment are safe and that Canada meets its international obligations.

NSCA

The NSCA was passed by Parliament on 20 March 1997, and became law in May 2000. This was the first major overhaul of Canada's nuclear regime since the Atomic Energy Control Act (AECA) and the creation of the Atomic Energy Control Board (AECB) in 1946. The NSCA incorporates stringent regulations to ensure that public health and safety are protected and is the key piece of legislation that ensures the safety of the nuclear industry and RAW management in Canada. The Canadian Nuclear Safety Commission (CNSC), established under the NSCA, is Canada's independent nuclear regulatory body. The CNSC comprises the Commission Tribunal, which makes licensing decisions, and the CNSC's staff organization, which prepares recommendations to the Commission Tribunal, exercises delegated licensing and authorization powers and assesses licensee compliance with the NSCA, the Act's associated regulations and licence conditions. The NSCA gives the CNSC the power to make regulations. Its mission is to regulate the use of nuclear energy and materials to protect health, safety, security and the environment, and to implement Canada's international commitments on the peaceful use of nuclear energy.

Regulations

There are nine safety-related regulations issued under the NSCA:

1. General Nuclear Safety and Control Regulations
2. Radiation Protection Regulations
3. Class I Nuclear Facilities Regulations
4. Class II Nuclear Facilities and Prescribed Equipment Regulations
5. Uranium Mines and Mills Regulations
6. Nuclear Substances and Radiation Devices Regulations

7. Packaging and Transport of Nuclear Substances Regulations
8. Nuclear Security Regulations
9. Nuclear Non-proliferation Import and Export Control Regulations

The CNSC's regulatory framework consists of regulations, policies, standards and guides that apply to all nuclear industries.

CNSC regulatory documents

The NSCA and its associated regulations provide the basis for regulatory expectations and decisions. Regulatory documents clarify NSCA requirements and associated regulations, and are an integral part of the regulatory framework for nuclear activities in Canada. Each regulatory document aims to disseminate objective regulatory information to stakeholders, including licensees, applicants, public interest groups and the public, and promote consistency in the interpretation and implementation of regulatory requirements. As outlined in the CNSC Regulatory Policy P299, *Regulatory Fundamentals* (CNSC, 2005), CNSC sets requirements using appropriate industry, national and international standards. The CNSC regulatory framework draws upon Canadian and international standards and best practices, including the nuclear safety standards of the International Atomic Energy Agency (IAEA).

A list of CNSC's regulatory documents is available online at: nuclearsafety.gc.ca. Two of these documents are specific to the management of RAW. Other more generic regulatory documents that relate to action levels, decommissioning, environmental protection and public information programs may also apply to the management of RAW. The CNSC's regulatory documents for management of radioactive waste are discussed below.

The CNSC Regulatory Policy P-290, *Managing Radioactive Waste* (CNSC, 2004) outlines the philosophy and principles used by the CNSC in regulating radioactive waste. The policy considers the extent to which owners of RAW must address:

- waste minimization;
- the radiological, chemical and biological management of RAW;
- the predicted impacts on the health and safety of persons and the environment;
- the measures needed to prevent unreasonable risk to both present and future generations; and
- the trans-border effects on the health and safety of persons and the environment.

The CNSC Regulatory Guide G-320, *Assessing the Long Term Safety of Radioactive Waste Management* (CNSC, 2006) assists licensees and

applicants to assess the long-term storage and disposal of RAW. The guide was developed using provincial, federal and international documents, following a consultation with the nuclear industry in Canada.

In addition, the nuclear industry in Canada, in conjunction with the CNSC, has developed two Canadian Standards Association (CSA) standards for the interim management of used nuclear fuel and RAW. These standards incorporate best practices both nationally and internationally. For example, the CSA has developed a standard consisting of best practices for the safe siting, design, construction, commissioning, operation and decommissioning of facilities and associated equipment for the dry storage of irradiated fuel, known as CSA N292.2-07, *Interim Dry Storage of Irradiated Fuel*. (CSA, 2007). The standard CSA N292.3-08, *Management of Low- and Intermediate-Level Radioactive Waste* (CSA, 2008) provides advice on the management of low- and intermediate-level radioactive waste which is based on current best practices, international experience and guidance, and in accordance with the existing CNSC regulatory requirements.

Regulatory approach

The Canadian regulatory approach to the safety of RAW management is based on three principles: life cycle responsibility and licensing, in-depth defence, and multiple barriers.

The CNSC uses a comprehensive licensing system for the management of radioactive waste, which is regulated during its entire life cycle – from site preparation, construction and operation to decommissioning and, finally, abandonment. It is the licensee's responsibility to demonstrate that a facility for RAW management can and will be operated safely throughout the lifetime of the facility. This step-wise approach requires a separate licence at each phase. The CNSC also requires early planning, as the application must submit all the relevant information associated with the site operation and decommissioning plans and financial assurance at the first stage. The outcome of the licensing process feeds back into the compliance program – to verify that the licensee fulfills the regulatory requirements.

The CNSC utilizes a harmonized or joint review approach with other federal, or provincial or territorial departments in such areas as health, environment, transport and labour. This approach allows for participation in the CNSC's assessment, licensing and compliance programs for waste management facilities.

International

Canada must also demonstrate how it continues to meet the obligations under the terms of the *Joint Convention on the Safety of Spent Fuel*

Management and on the Safety of Radioactive Waste Management (IAEA, 1997). The Joint Convention is based on IAEA guidelines and standards. The Joint Convention is an international agreement, governing all aspects of nuclear fuel waste and radioactive waste management. Therefore, for the management of used nuclear fuel, uranium mines and mills and RAW, facilities must be designed, operated, and decommissioned in order that Canada can demonstrate it meets the obligations of the Joint Convention.

19.1.3 Waste classification

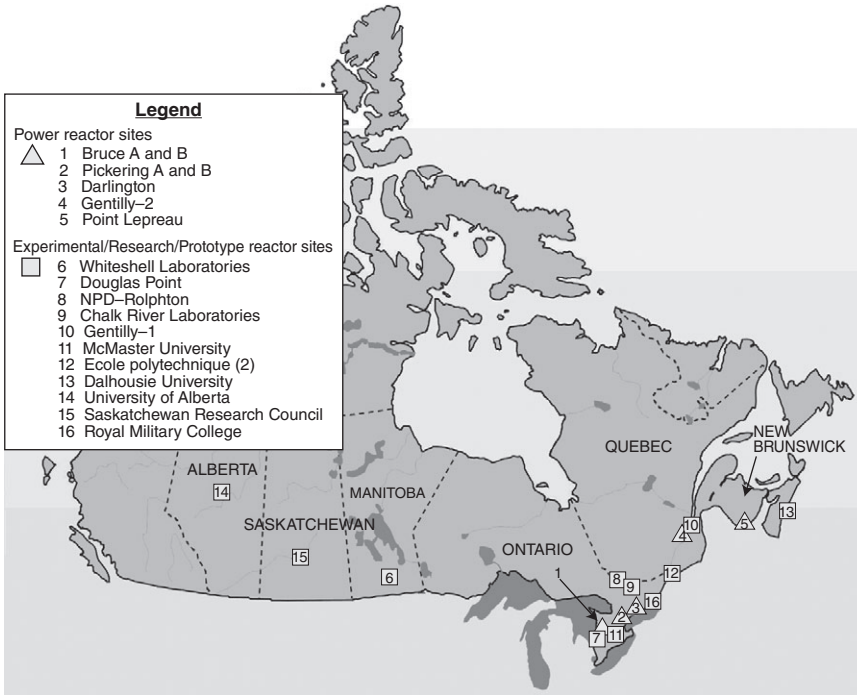
The radioactive waste classification system in Canada is based on the CSA standard, CSA 292.3-08 (CSA, 2008), which takes into account the IAEA Safety Guide SSG-1, *Classification of Radioactive Waste* (IAEA, 2009), and the needs of the Canadian industry. The system recognizes four main classes of radioactive waste:

1. high-level radioactive waste (HLW),
2. intermediate-level radioactive waste (ILW),
3. low-level radioactive waste (LLW), and
4. uranium mine and mill waste.

The waste classification system is organized according to the degree of containment and isolation required to ensure safety in the short and long term. It also takes into consideration the hazard potential of different types of RAW.

19.2 Radioactive waste (RAW) management strategies

In keeping with the *Policy Framework for Radioactive Waste* (Natural Resources Canada, 1996), Canada has taken different approaches for the management of high-level radioactive waste (i.e., used nuclear fuel), low- and intermediate-level radioactive waste and uranium mine and mill tailings. These approaches reflect not only the different characteristics of the wastes, but also the economics and the geographic dimensions of Canada and the locations of the waste. Figure 19.1 (LLRWMO, 2009) shows the locations of nuclear sites in Canada. Canada's national strategy on radioactive waste management includes a national approach for the long-term management of high-level radioactive waste, regional solutions for low and intermediate-level radioactive waste, and the long-term management of uranium mining and milling waste close to uranium mine and mill sites. Canada does not have a central, national waste disposal facility for radioactive waste. Currently, all waste is in safe storage or is disposed on site (e.g., uranium mine tailings).



19.1 Nuclear sites in Canada (LLRWMO, 2009).

19.2.1 Nuclear power utilities

Used fuel

There are 22 CANDU[1] (CANada Deuterium-Uranium) power reactors in Canada owned by three provincial electric utilities. Ontario Power Generation Inc. (OPG) owns 20 reactors (eight of which are leased to Bruce Power Inc. for commercial electricity production), while Hydro-Québec (HQ) and New Brunswick Power (NBP) each own one reactor. All CANDU® fuel bundles are fabricated from natural uranium oxide pellets, contained in a zirconium-alloy (Zircaloy-4) sheath. The weight of a nominal bundle is 23.6 kg, of which 21.3 kg is due to the uranium oxide, approximately 19.2 kg can be attributed to the uranium (without the oxygen component). Each year, 4,500–5,400 used fuel bundles are generated per reactor, based on 80–95% full power reactor operation (CNSC, 2008). A 600 MW CANDU® nuclear reactor produces approximately 20 m^3 of used nuclear fuel per year.

Nuclear fuel wastes from nuclear power generating stations are stored in wet and dry states at the locations where they are generated. The used fuel

[1]CANDU® is a registered trademark of Atomic Energy of Canada Limited.

is first placed in water-filled fuel storage bays, and after several years (i.e., six to ten years) the used fuel can be transferred to an on-site dry storage facility. These dry storage facilities are large, reinforced concrete cylinders or containers. Each nuclear power generating station in Canada has enough storage space to store all the used fuel produced during the operating life of the station.

Following a decade-long environmental assessment of a deep geological disposal concept for nuclear fuel waste that ended in 1998, the Government of Canada passed the NFWA in 2002, which made owners of nuclear fuel waste responsible for the development of long-term waste management approaches. Shortly after the NFWA came into force, the nuclear energy corporations, OPG, HQ and NBP, established the NWMO and each waste owner established trust funds to finance the implementation of long-term waste management activities. The NWMO's mandate is to explore options for the long-term management of Canada's nuclear fuel waste, provide proposals to the Government of Canada and to implement the selected approach.

Following extensive studies and public consultation, the NWMO presented four options, including those listed in the NFWA, namely long-term storage at the reactor sites, central shallow or below ground storage, deep geological disposal, and lastly an option called the adaptive phased management (APM) approach (NWMO, 2005). The APM approach essentially combines the three above listed options within a flexible adaptive management decision-making process. In 2007, the Government of Canada announced that it had selected the APM approach for the long-term management of used fuel in Canada. With this government decision, NWMO assumed responsibility for implementing the APM approach (NWMO, www.nwmo.ca).

Low- and intermediate-level waste

In Canada, the nuclear energy corporations (i.e., OPG, HQ and NBP) are responsible for the long-term management of RAW generated from their nuclear reactor operations. Currently, the LLW and ILW are stored in a variety of structures located in waste management facilities at nuclear power generating stations. Prior to storage, the volume of the wastes may be reduced by incineration, compaction or shredding. In addition, within the stations there are facilities designated for the decontamination of parts and tools, laundering of protective clothing and the refurbishment and rehabilitation of equipment.

The LLW and ILW from OPG's nuclear power generating stations are stored on an interim basis at the Western Waste Management Facility (WWMF) adjacent to the Bruce Nuclear Power Development (BNPD),

which is located in the municipality of Kincardine, Ontario. In April 2002, OPG and the Municipality of Kincardine signed a Memorandum of Understanding to jointly study options for the long-term management of the wastes at the WWMF. Following a strong positive response from local residents, OPG proceeded in 2005 with plans to construct a deep geological repository (DGR) at a depth of 680 m for its low- and intermediate-level wastes near the BNPD. The DGR will be designed to hold OPG's current and future LLW and ILW from its 20 CANDU® reactors (OPG, http://www.opg.com/power/nuclear/waste/dgr/index.asp).

19.2.2 Nuclear research and test establishment facilities

There are two main nuclear research facilities in Canada: Atomic Energy of Canada (AECL) – Chalk River Laboratories (CRL), located in Chalk River, Ontario is operational, and AECL – Whiteshell Laboratories (WL), located in Pinawa, Manitoba is undergoing decommissioning. AECL is responsible for the long-term management of RAW generated by CRL, WL and the three partially decommissioned prototype reactors (i.e., Douglas Point, Gentilly-1 and Nuclear Power Demonstration, NPD), as well as for the low- and intermediate-level waste it accepts from off-site waste generators on a fee-for-service basis. AECL is also responsible for managing its used fuel, including research reactor fuel and any used CANDU® fuel sent to its laboratories for examination, until the NWMO is ready to accept the waste for management in facilities constructed under the APM approach.

In 2006, the Government of Canada adopted a new long-term (70-year) strategy to deal with the nuclear legacy liabilities that have resulted from over 60 years of nuclear research and development carried out on its behalf at AECL sites. The overall objective of the long-term strategy is to safely and cost-effectively reduce the liabilities and associated risks based on sound waste management and environmental principles in the best interests of Canadians. The Nuclear Legacy Liabilities Program (NLLP) was established in 2006 and is being implemented through a Memorandum of Understanding between Natural Resources Canada (NRCan) and AECL (NLLP, www.nuclearlegacyprogram.ca; Miller *et al.*, 2008; Metcalfe *et al.*, 2009). AECL's ongoing LLW and ILW will be dealt with in waste management facilities that will be built under the NLLP.

At other nuclear research sites, RAW materials are segregated by licensees into short-lived and long-lived RAW. Short-lived RAW is stored on-site to allow for decay until it can be disposed of in a conventional manner. Long-lived RAW is kept on-site temporarily until a certain amount or volume is accumulated; thereafter it may be sent off site using a commercial service provider, as available, or transported to AECL-CRL for safe storage, also under a fee-for-service basis.

As of March 2011, there are seven operating research reactors in Canada (see Fig. 19.1). In the past, research reactors have typically used highly enriched uranium (HEU) fuel, obtained from the United States for the fuel cores. Within the last decade, some of the cores have been converted to low-enriched uranium (LEU) fuel as part of the Global Thread Reduction Initiative (see Section 1.4.3). The used fuel from the research reactors is either sent to AECL-CRL for storage or, in the case of HEU, returned to the United States for processing.

19.2.3 Fuel manufacturing

General Electric Canada Incorporated and Zircatec Precision Industries Incorporated are the only fuel fabricators in Canada. Fuel manufacturing waste consists of a variety of potentially uranium-contaminated wastes, and following implementation of varying recycling initiatives, the residual volume of LLW is drummed and stored in warehouses pending the establishment of an appropriate long-term waste management facility.

19.2.4 Uranium mines and mill facilities

Canada's operating uranium mining companies, Cameco Corporation and Areva Resources Canada Incorporated are not only leaders in uranium production, but they also lead in the development of environmentally sustainable uranium mining practices. They have developed new technologies to manage uranium mill tailings and reduce environmental impacts.

The tailings management strategy is based on two principles that underlie the containment of the tailings and their potential radionuclide and heavy metal contaminants:

1. Hydraulic containment during the operational phase: The pit is maintained in a partially dewatered state throughout the operational life of the tailings facility to create a cone of depression in the groundwater system, which results in the natural flow being directed toward the pit from every direction. Since water has to be pumped continuously from the pit, current water treatment technology results in high-quality effluent suitable for discharge to surface water.
2. Passive long-term containment, using the hydraulic conductivity contrast between the tailings and their surrounding geologic materials: Long-term environmental protection is established through control of the tailings' geochemical and geotechnical characteristics during tailings preparation and placement. This control creates future passive physical controls for groundwater movement in the system, which will exist after the decommissioning of operational facilities.

In addition to tailings from the milling process, uranium mining results in large quantities of waste rock being produced. The segregation of these

materials according to their future management requirements is now a core management strategy. Material excavated from open pits is classified into three main categories: clean waste (both overburden and waste rock), special waste (containing sub-economic mineralization) and ore.

The clean waste refers to waste materials that are benign with respect to future environmental impact, and that can be disposed of in surface stockpiles or used on-site for construction purposes. The special waste is waste rock near ore bodies. This waste is potentially problematic, because it has some halo mineralization around the ore deposit, and is therefore potentially acid-generating in some instances and/or a source of contaminated leachates when exposed to an oxidation environment. The disposal of this special waste in mined-out pits and flooding, to cut off the oxygen supply from the atmosphere and stop oxidation reactions, is now a widely recognized solution. If the pit is not suitable for the long-term management of the risk, engineered covers present an *in-situ* solution to impede the interaction of oxygen and moisture with the special waste. Typically, any waste material with uranium content greater than either 300 ppm U_3O_8 or 0.025% (250 ppm) uranium is categorized as special waste, and all material grading greater than 0.085% uranium has been classified as ore. The cut-off grade for the mill may vary depending on market conditions for uranium.

All mine and mill facilities provide water treatment systems to manage contaminated water collected from their tailings' disposal facilities, as well as water inflows collected during open pit or underground mining, and problematic seepages from waste rock piles. The treatment processes vary from continuous to batch systems, and largely rely on conventional physical settling and chemical precipitation methods found in the metal mining industry.

Owners of closed uranium mines are also required to ensure that their sites are properly decommissioned, and that they have set the standard for decommissioning uranium mine sites. In instances where remedial actions are required at uranium mine and mill tailings facilities where the owner no longer exists, the Government of Canada and provincial governments ensure that the sites are safely decommissioned through cost-sharing arrangements.

19.2.5 Historical waste

Historical low-level RAW in Canada refers to LLW that was managed in the past in a manner no longer considered acceptable, but for which the current owner cannot reasonably be held responsible, upon which the Government of Canada has accepted the long-term responsibility. In 1982, the Government of Canada established the Low-Level Radioactive Waste Management Office (LLRWMO) within AECL as the federal agent for the

cleanup and management of historical LLW in Canada. While NRCan provides policy direction and funding, the LLRWMO develops and implements the Government of Canada's strategic approach to historical waste management by working with communities and federal stakeholders to develop solutions to safely and cost-effectively reduce liabilities and associated risks (LLRWMO, www.llrwmo.org).

The LLRWMO has completed historical waste cleanups across Canada and continues to monitor several sites with historic radium or uranium contamination. At some sites, materials have been placed in interim storage pending the development of a long-term management approach. Ongoing site monitoring, inspection and maintenance are conducted at these sites.

19.2.6 Radioisotope production and use

Radioisotope production and use generate a variety of radionuclides for commercial use, such as cobalt-60 for sterilization and cancer therapy units, and molybdenum-99 or other isotopes for use as tracers for medical research, diagnoses and therapy. Wastes that are generated during production are managed by the respective producers.

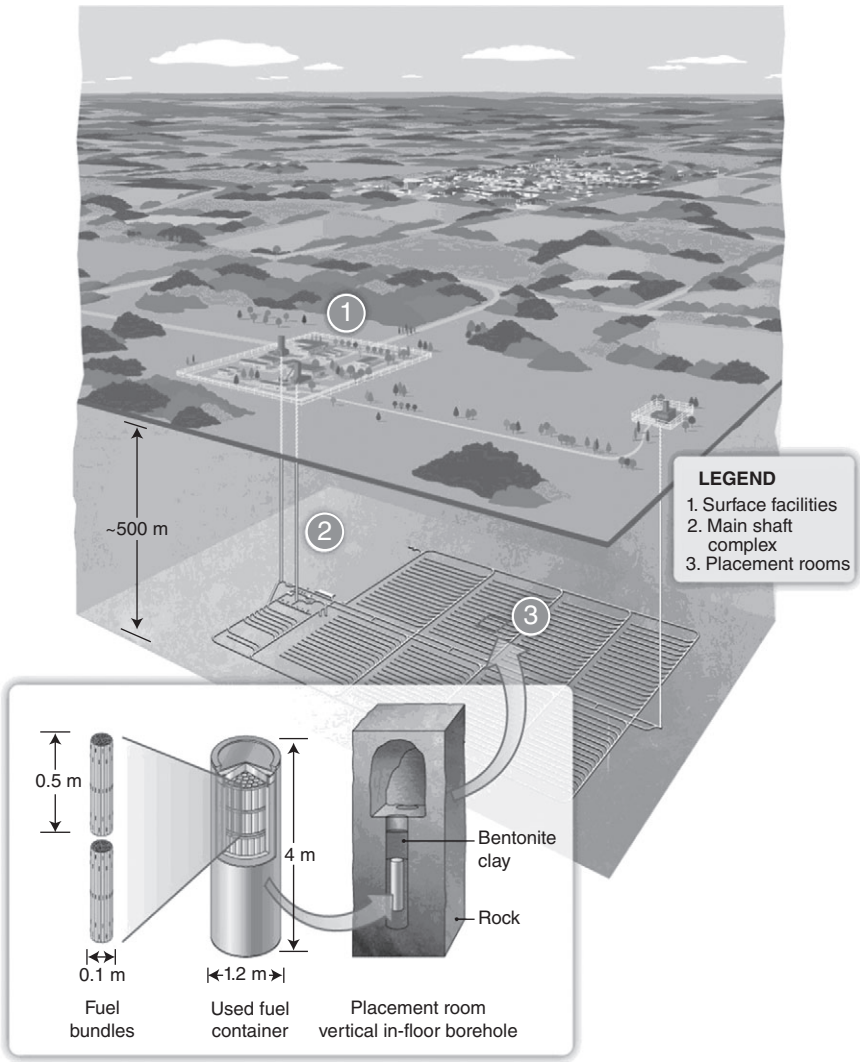
A number of waste management facilities process and manage the wastes that result from the use of radioisotopes for research and medicine. In general, these facilities collect and package waste for shipment to approved storage sites such as AECL-CRL. In some cases, the waste is incinerated, or retained in the facility to allow for decay to insignificant radioactivity levels and then released as clean materials.

19.3 Long-term management

19.3.1 Used fuel

The selected and approved NWMO's APM approach for long-term management of Canada's used fuel comprises both a technical method and a management approach (NWMO, 2005). The technical method is based on centralized containment and isolation of the used fuel in a deep geological repository in a suitable rock formation (Fig. 19.2). It provides for continuous monitoring of the used fuel and the potential for retrievability for an extended period of time. Consistent with adaptive management, there is provision for contingencies, such as the optional step of shallow storage at the selected central site if circumstances favour early centralization of the used fuel before the repository is ready.

The management system is based on phased and adaptive decision making. Flexibility in the pace and manner of implementation allows for phased decision making, with each step supported by continuous learning,



19.2 Conceptual deep geological repository for nuclear fuel waste (NWMO, 2005).

research and development, and public engagement. An informed, willing community will be sought to host the centralized facilities. Sustained engagement with people and communities is a key element of the plan, as the NWMO continues to work with all stakeholders (i.e., citizens, communities, municipalities, all levels of government, Aboriginal organizations, industry and others).

NWMO's implementation activities within its initial five-year plan are focusing on seven key areas:

1. building a relationship with key stakeholders,
2. site selection,
3. design and safety case for APM deep geological repository,
4. financial surety,
5. adapting plans,
6. accountability and governance, and
7. building the organization.

19.3.2 Low-and intermediate-level radioactive waste

The owners of low- and intermediate-level radioactive waste are managing and operating storage facilities for their wastes. In addition, the two major waste owners, OPG and AECL, are pursuing initiatives to develop and implement long-term management solutions.

Long-term management of OPG's low- and intermediate-level radioactive waste

Ontario Power Generation Inc. has recognized that, while its current approach to RAW storage is safe, secure, and environmentally sound, a new approach will be required for the long-term management. As described in Section 19.2.1, under the terms of a Memorandum of Understanding, OPG and Kincardine engaged a consulting firm to conduct an independent assessment study (IAS) of the feasibility, safety, social and economic feasibility and the potential environmental effects of a proposed long-term management facility at the WWMF. Three options were studied: enhanced processing and storage, a covered above-ground concrete vault and a deep geologic repository. The IAS concluded that each of the options were feasible. The options could be constructed to meet international and Canadian safety standards with a high margin of safety. In April 2004, Kincardine Council passed a resolution to select the 'Deep Rock Vault option as the preferred course of study' for the management of LLW and ILW because it had the highest margin of safety and is consistent with best international practice.

The DGR involves the construction of rock vaults within stable, low permeability bedrock using conventional mining techniques. The rock vaults would be positioned at a depth of approximately 680 m in relatively flat-lying sedimentary rock formations that have remained tectonically stable and undeformed for hundreds of millions of years. Access to the repository would be through a vertical, concrete-lined shaft. A second shaft would be constructed for ventilation and emergency egress purposes.

In December 2005, OPG submitted a letter of intent to construct the DGR to the CNSC, thus initiating the environmental assessment (EA)

process. Following detailed geo-scientific investigations, preliminary design work and environmental and safety assessments, the Environmental Impact Statement, Preliminary Safety Report and supporting reports were submitted to CNSC in March 2011 (OPG, http://www.opg.com/power/nuclear/waste/dgr/index.asp). EA approval and a site preparation and construction licence are expected to be received in 2012/2013 and the earliest in-service is expected in 2018.

Long-term management of AECL's low and intermediate-level radioactive waste

As described in Section 19.2.2, the Government of Canada is implementing a long-term strategy, the NLLP, to deal with the nuclear legacy liabilities at AECL sites. The program addresses environmental restoration, infrastructure decommissioning, waste management, and care and maintenance of the nuclear liabilities until they are addressed.

An important element of the program is the development of an integrated waste plan to ensure the selection of the optimal mix of enabling facilities and their implementation schedules to address the current and future wastes arising from the program's activities. An interim integrated waste plan for CRL has been developed, and will be expanded to the other sites and go through multiple iterations of refinement as additional planning and waste characterization data are obtained.

As input to the refinements of the integrated waste plan, a number of studies are under way to better define the waste processing, treatment and long-term management facilities required to deal with the wide variety of legacy waste types at AECL sites. This will help to define, for example, the volume reduction and waste immobilization technologies to be used, the extent to which buried waste can be managed in place over the long term, and the available options for the long-term management of the waste that needs to be recovered.

Significant savings in long-term waste management costs at CRL could be achieved by constructing a very low level waste (VLLW) facility to receive large volumes of VLLW wastes such as soil, concrete, vegetation, asphalt and/or building materials/rubble that are being generated by NLLP infrastructure decommissioning and environmental remediation projects and activities. All pre-project activities have been completed to support the development of a VLLW facility, and formal project development activities are being initiated.

In 2006, an investigation was initiated to assess the feasibility of the bedrock at the CRL site to host a proposed geological waste management facility (GWMF) as a final enabling facility for the long-term management of low- and intermediate-level solid radioactive waste at the site.

The feasibility study involved exploring the geoscience and engineering characteristics of the proposed bedrock and the drilling and testing of characterization boreholes. The collected information and interpretations were then used to construct three-dimensional deterministic computer models of the geology of the site and the associated groundwater flow regime. The results of the feasibility study are currently under review by various parties to assist in the decision-making process for proceeding.

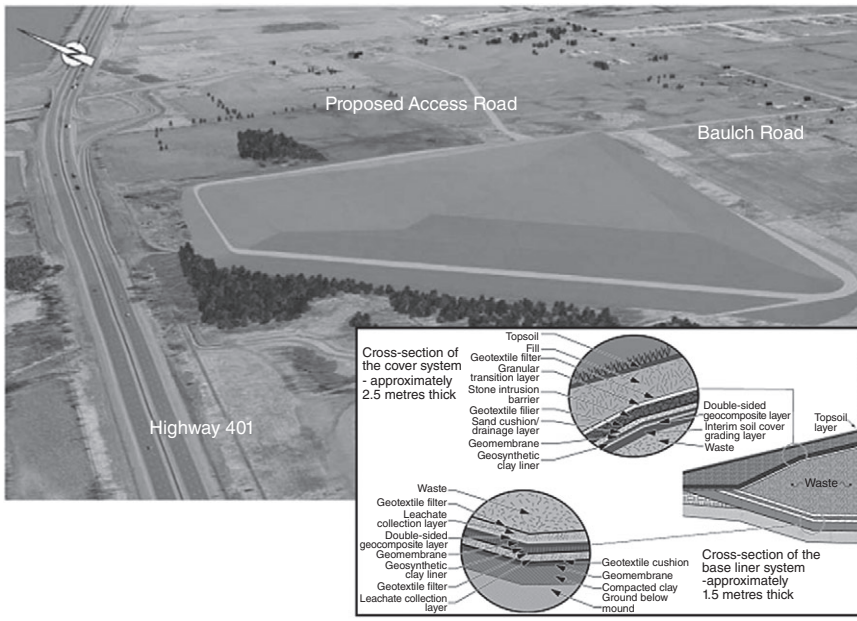
AECL has initiated discussions with the NWMO on the long-term management of AECL's varied inventory of legacy research reactor fuel waste within the NWMO APM approach.

19.3.3 Historical waste

The bulk of Canada's historical LLW is located in the southern Ontario communities of Port Hope and Clarington. These wastes and contaminated soils amount to roughly 1.7 million m^3. They originate from the operations of a radium and uranium refinery in the municipality of Port Hope, dating back to the 1930s. While recognizing that there are no urgent risks from a health or environmental standpoint, the Government of Canada determined that intervention measures are required in order to implement more appropriate long-term management measures for these materials.

In March 2001, the Government of Canada and local municipalities entered into an agreement on community-developed proposals to address the cleanup and long-term management of these wastes, thereby launching the Port Hope Area Initiative (PHAI) with two projects: Port Hope and Port Granby. The PHAI will result in the long-term management of these historical wastes in two above-ground mounds (Fig. 19.3) that will be constructed in the local communities (PHAI, www.phai.ca; Fahey and Case, 2010). AECL is the proponent for the PHAI on behalf of the Government of Canada and, in conjunction with NRCan and Public Works and Government Services Canada, have formed the PHAI Management Office (PHAI MO) to plan and manage the overall PHAI execution.

The Port Hope Project entails the cleanup of the urban area and 14 major sites and the consolidation of all of the wastes (approximately 1.2 million m^3) in the municipality of Port Hope at one long-term waste management facility (WMF). This facility is to be located at the present site of the existing Welcome Waste Management Facility. The wastes consist of LLW and contaminated soils, containing radium-226, uranium and arsenic as the primary contaminants. The LLW in interim storage and contaminated soils from various sites, including the Port Hope harbour, will be excavated and safely transported to the new long-term WMF. For selected sites, specific methods will be required for waste excavation and removal. For example, the contaminated sediment in the Port Hope harbour will be removed by dredging,



19.3 Port Hope mound design.

dewatered in geo-synthetic containment tubes and transported to the long-term WMF for storage (Case and Kolberg, 2011).

The Port Granby Project involves the relocation of the existing Port Granby wastes (approximately 0.5 million m^3) to a new above-ground, long-term WMF. The WMF is to be located at a nearby site approximately 700 m north of the current site and away from the Lake Ontario shoreline.

The cleanups in Port Hope and Port Granby are anticipated to be completed by 2020. Following the emplacement of wastes and the closure of the new WMFs, the long-term monitoring and maintenance phase will commence and continue for hundreds of years.

19.4 Contaminated site cleanup experience and planned projects

19.4.1 Low-level, historical waste projects

A variety of sites contaminated with historical low-level radioactive waste materials have been identified across Canada. The diversity of wastes, wasteforms and sites include: pitchblende ore handling facilities used during the 1930s, 1940s and 1950s along a 2,200 km transportation route between the Port Radium mine in the Northwest Territories and Fort McMurray in

northern Alberta; uranium and radium processing residues currently located in Port Hope area waste sites established during the 1930s, 1940s and 1950s; discarded luminescent dials and apparatus found at sites across Canada; and former radium dial painting and waste management operations located in the Toronto area.

As described in Section 19.2.5, the Government of Canada established the LLRWMO in 1982 to characterize and delineate these historical low-level waste sites across Canada, and undertake decontamination, waste consolidation and interim waste storage as required at these sites. The types of remedial work conducted by the LLRWMO at these historical waste sites include: excavation and transportation of radioactively contaminated soil in quantities ranging from a few m^3 to thousands of m^3; collection/consolidation of contaminated debris and radioactive artifacts; decontamination of residential and industrial structures primarily associated with historical radium dial painting operations; and the development of community-based interim waste management solutions pending the development of a longer-term solution. This work routinely involves liaison with the local communities and regulatory agencies to develop acceptable waste management solutions for the short and long term (Benitez *et al.*, 2011; Gardiner *et al.*, 2011).

Most of the remaining historical waste to be dealt with in Canada is located along the northern transportation route. The waste has resulted from the past transport of radium and uranium-bearing ore and concentrates from the Port Radium mine to the barge-to-rail transfer point at Fort McMurray. The sites that still have to be remediated include Sawmill Bay, Bennett Landing, Bell Rock and Fort Fitzgerald. Strategies are currently being developed for the cleanup of these remaining sites. They are estimated to consist of about 10,000 m^3 of contaminated soils.

19.4.2 Uranium mine and mill tailings management areas

There are 20 tailings management sites that have resulted from the operation of uranium mines in Canada: 14 in Ontario, four in Saskatchewan and two in the Northwest Territories. Decommissioning of uranium mines and mills is governed by the Uranium Mine and Mills Regulations under the NSCA. The Cluff Lake Project is described here as an example of the type of activities that are undertaken for safe decommissioning.

Cluff Lake project

The Cluff Lake Project, which is owned and operated by AREVA, was completed at the end of 2002, when ore reserves were depleted. More than 28 million tonnes of U_3O_8 was produced over the 22-year life of the project.

Site facilities included the mill and tailings management area (TMA), four open-pit and two underground mines, the camp for workers and site infrastructure. Cluff Lake was the first of the northern Saskatchewan uranium mines to be decommissioned. The decommissioning licence was received from the CNSC in July 2004. The objective is to return the site as closely as practical to its original state in a manner that both protects the environment and allows traditional uses such as fishing, trapping and hunting to be carried out safely.

Decommissioning the mill involved two phases, which were completed in 2004 and 2005. The mill demolition work was broadly similar to demolition of other comparable size industrial facilities, with special measures needed to protect workers from residual contamination and industrial hazards, and to prevent the spread of contaminants into the environment. Waste materials were disposed of in one of the open pits at the site, together with much larger volumes of waste rock.

Decommissioning of the TMA was initiated by covering the tailings with till[2] in stages to promote consolidation. The local till material developed from an adjacent borrow area was used for covering the tailings materials. When consolidation was complete, the TMA cover was contoured to provide positive drainage, using locally available till with a minimum cover thickness of 1 m, and then re-vegetated. Extensive characterization of the tailings and the site's geology and hydrogeology has been performed to acquire reliable data on which to base the assessment of long-term performance. One of the objectives of the follow-up monitoring program is to verify the key assumptions used in the long-term performance assessment.

Two open pits have been used for the disposal of waste rock, with one of these two pits also used to accept industrial waste during operations and decommissioning. This waste included the mill demolition waste.

19.4.3 Chalk River Laboratories long-term strategy

AECL's CRL site is large (~4000 ha) and diverse and contains many structures and features, some dating back to the beginning of the site's first establishment in 1944. The site is expected to continue in operation as a licensed nuclear site, with a wide range of nuclear research and development and operation activities being conducted for many years to come. Any contaminated facilities or contaminated lands and radioactive wastes that have been produced during prior operations or decommissioning activities

[2]Till or glacial till is unsorted glacial sediment. Glacial drift is a general term for the coarsely graded and extremely heterogeneous sediments of glacial origin. Glacial till is that part of glacial drift which was deposited directly by the glacier. Its content may vary from clays to mixtures of clay, sand, gravel and boulders.

constitute the legacy liabilities that are now managed through the NLLP, described above in Section 19.2.2.

The decommissioning model for the CRL site, including the waste management areas, is described in a comprehensive preliminary decommissioning plan (CPDP) (Miller, 2010). The strategy developed is for a number of individual decommissioning projects for the site's various components over time rather than a single project for the site as a whole at some time in the future, designated as the end of operational life. Priorities for decommissioning projects are established based on health, safety, security and environmental risks, and also take into consideration operational requirements and business priorities (Stephens, 2009). The individual decommissioning projects in the CRL site CPDP document are grouped into seven planning envelopes (PE), where each PE is a grouping that has a degree of similarity, which lends itself to the application of common planning assumptions. Planning envelopes 1–4 are for above-ground structures, PE 5 is for distributed services, PE 6 is for affected lands, and PE 7 is for waste management areas. The individual projects will, in general, take each respective structure or feature to a documented end-state while the site as a whole continues in operation. Some projects will be implemented at the end of the site's operational life to qualify the site for a period of institutional control, the reference being 300 years (2100–2400). During the institutional control period, selected parts of the site may be turned over for industrial re-use in accordance with then-current laws and regulations. Work is currently under way to develop an overall, co-ordinated environmental restoration strategy for the CRL site which is integrated with the plans for decommissioning the physical structures and available waste management facilities, and which is in the most cost- and risk- effective manner as determined by the various stakeholders.

19.4.4 Whiteshell Laboratories long-term strategy

Whiteshell Laboratories (WL) has provided research facilities for the Canadian nuclear sector since the early 1960s. In 1997, AECL decided to discontinue research programs and operations at the facility, and in 1999 began to prepare plans for the safe and effective decommissioning of the WL site.

The major structures located on the WL site include the organic-cooled WR-1 research reactor (in storage with surveillance since 1995), the shielded facilities, research laboratories, and liquid and solid RAW management areas and facilities, including the concrete canister storage facility for the dry storage of research reactor fuel. In preparation for decommissioning, a comprehensive environmental assessment was successfully completed (AECL, 2001), and the CNSC issued a decommissioning licence for the WL site which came into effect on 1 January 2003. The CNSC has approved a

detailed decommissioning plan for the site, which provides information, as required, under the Class I Nuclear Facilities Regulations.

Initially, the decommissioning activities have been focused on decontaminating and modifying nuclear facilities, laboratories and the associated service systems and removing redundant buildings to reduce risk and operating costs. As buildings are prepared for final decommissioning, enabling facilities are being constructed to handle and store contaminated wastes that will arise from decommissioning (Koroll *et al.*, 2009). Other work is ongoing in support of commitments made during the environmental assessment, including ongoing confirmation of the hydrogeological conditions of the waste management area on the site, fitness-for-service studies and an updated groundwater monitoring plan. Some areas within the waste management area were identified for early remediation within the overall site plan, including the fuel-bearing (in-ground) standpipes (Stepanik *et al.*, 2011). The current plan has the major nuclear facilities being finally decommissioned/dismantled in the 2020–2035 timeframe, consistent with the planned availability of final long-term waste management facilities. These plans undergo reassessment as new cost and schedule information becomes available. It is anticipated that the site will be under institutional control for an extended period following decommissioning of the site infrastructure.

19.5 Case studies and lessons learned

19.5.1 Whiteshell laboratories decommissioning

As discussed above, the Whiteshell Laboratories decommissioning project to date has focused on decontaminating and modifying nuclear facilities, laboratories and the associated service systems and removing redundant buildings to reduce risk and operating costs. The lessons learned associated with the management of the waste already stored on the site and produced during the decommissioning activities arise from three main activities: job planning, physical decommissioning, and maintaining safety through the decommissioning project timeline.

The waste management strategy must be developed in advance, with the flow of waste materials and the required resources identified to ensure that 'waste material flow' does not become the critical path, and limit the progress of the physical decommissioning. The process used involved radiologically screening and segregating the waste at the source. This task can be long and repetitive; therefore, it was found beneficial to rotate workers to enhance training of staff and mitigate human error. To enhance waste material flow in some situations, a best practice 'lean manufacturing' philosophy was applied to the development of the material handling and monitoring process. This planning philosophy had the added benefit of reducing overall

costs for the activity, as well as maintaining a high level of worker morale as there were limited bottlenecks in the activity. There is a requirement to ensure the safety of the physical structures within the WL waste management areas from a variety of perspectives, including upkeep to today's standards and the development of plans for waste removal and transfer to a long-term waste management facility. Key lessons from the work to date include the importance of records, the need for ongoing geotechnical assessment of the area and in-ground structures, and the need for technology for assessment and characterization prior to developing and executing work plans to ensure worker safety throughout any planned waste retrieval. This latter work is in its early stages and interaction with other international groups is assisting with the development of safe work plans.

19.5.2 Port Hope Area Initiative

The Port Hope Area Initiative 2001 Legal Agreement (Section 19.3.3) formalized a long-term, community-based solution for the long-standing issue of dealing with the contaminated soils within licensed, interim storage facilities and on municipal and private properties in the community of Port Hope itself. While the PHAI is ultimately a major environmental remediation project, with the planned construction of two above-ground, engineered containment facilities for an estimated 1.7 million m^3 of low-level waste-contaminated soil and debris, it is the long-standing involvement of the municipalities and the general public in the various project activities that define this project, its implementation protocols, and ultimately will define its success. The project has been built on various stages of community engagement and empowerment, community partnering and volunteerism. This engagement of key stakeholders is supplemented by an extensive public communication and stakeholder outreach program. The Environmental Assessment follow-up program defines a number of requirements for significant project-specific monitoring activities and, as required, mitigation measures. To ensure the project effects will be understood, detailed plans have been produced to extensively monitor the predicted socio-economic and biophysical impacts. The project has reached this stage and will be successfully completed only with the continued cooperation of the host municipalities, the citizens, the regulators, and the implementers including the Government of Canada – an example in the making of true community and stakeholder engagement.

19.5.3 Northern transportation route

Currently, a key focus area for the LLRWMO is putting in place a strategy to address sites in Canada's north that were contaminated, long ago, by the

spillage of radioactive ores in transport. The contamination is located along what is known as the northern transportation route as shown in Fig. 19.4 (LLRWMO, www.llrwmo.org), a 2200 km route beginning at the former Port Radium site in the Northwest Territories and extending to northern Alberta. The LLRWMO is adapting methods that it has successfully used in Canada's southern regions. These methods of community engagement and technical approaches take into account the geography and the environment, while respecting the ways of inhabitants in the north. The success of the LLRWMO in southern communities has been based on building confidence with the communities involved through a carefully designed process, including cultivating stakeholder involvement early on in the process. Building and maintaining a community's confidence require constant commitment, significant resources and mutual effort. In northern communities, these engagement processes include taking into account traditional and local knowledge and providing training and outreach to ensure there is a strong community participation in the environmental remediation and

19.4 Northern transportation route in Canada (LLRWMO, www.llrwmo.org).

policy decision so that the community can participate in the stewardship of their natural environment. One of the challenges of such projects built on a participatory approach is the need to find and achieve a balance between stakeholder representation, stakeholder participation, and project progress and implementation. That balance can vary with the individual project and its stakeholders.

19.6 Acknowledgments

A key reference source for this chapter is the *Canadian National Report for the Joint Convention on the Safety of Spent Fuel Management and on the Safety of Radioactive Waste Management* – Fourth Report (Draft), to be published by the Canadian Nuclear Safety Commission. The authors of this chapter would like to acknowledge the efforts of the project team responsible for the development of this report – Julie Mecke, Anne McLay, Adelle Ferguson, Kathleen Hollington, Dale Huffman, Herminia Roman, Lisa Lang, and Pierre Wong.

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20

South Africa: experience of radioactive waste (RAW) management and contaminated site clean-up

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Abstract: This chapter describes the development of radioactive waste (RAW) management policies in South Africa and the implementation of such policies during contaminated site clean-up.

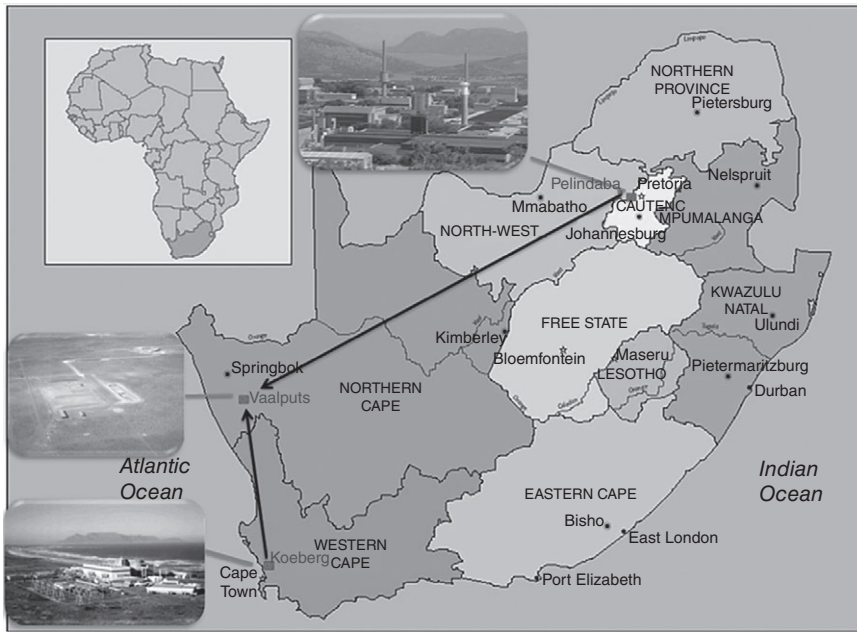
Key words: nuclear fuel cycle, nuclear waste, waste management, waste classification, nuclear reactor programme decommissioning.

20.1 Introduction

The main generators of solid radioactive waste (SRAW) in South Africa are the South African Nuclear Engergy Corporation (Necsa), Koeberg Nuclear Power Plant (NPP) and the mining industry. The other generators such as the iThemba Accelerator facilities, hospitals and industries are regarded as minor contributors (Fig. 20.1).

The South African nuclear programme of the 1970s to mid-1990s (mainly practiced at Necsa) has left the country with liabilities with regard to redundant, radioactively contaminated equipment, buildings and radioactive waste (RAW). RAW management policy in South Africa is structured and implemented by including the applicable sections from the various National Acts, i.e. National Environmental Management Act (No. 107 of 1998), The National Nuclear Regulator Act (No. 47 of 1999), the Nuclear Energy Act (No. 46 of 1999), the Hazardous Substances Act (No. 15 of 1973), and the National Water Act (No. 36 of 1998).

Radioactive wastes in South Africa are divided into two categories: historical waste and current/future waste. Historical radioactive waste, the main producer of which was Necsa, was generated prior to 1987. Necsa (South African Nuclear Energy Corporation) is a multi-facility nuclear site that operates or has operated the processes involved in the front-end of the nuclear fuel cycle (NFC) and therefore excludes the reprocessing of spent fuel (SF). The South African nuclear programme started in 1948 and focused on research and development in the NFC and in military applications. Some highlights in the history of Necsa are the successful separation of uranium



20.1 Locality map of South Africa showing Vaalputs, Koeberg and Pelindaba [1].

isotopes and the start of the uranium enrichment programme that included the uranium conversion facility (Fig. 20.2). The enriched uranium was used as fuel for the SAFARI-1 research reactor, the NPP at Koeberg and for military purposes.

The uranium conversion and enrichment research and production projects were terminated in the early 1990s, due to cost considerations. As stated above, the South African nuclear programme of the 1970s to mid-1990s has left the country with liabilities with regard to redundant, radioactively contaminated equipment and buildings and RAW. Necsa has been generating RAW since the commissioning of the SAFARI-I research reactor in 1965, and the waste includes fuel fabrication waste as well as uranium conversion and enrichment historical waste.

The bulk of Necsa's waste was, however, generated between 1970 and 1998 by the nuclear fuel production cycle, namely the uranium conversion, enrichment and fuel fabrication plants. The medical isotope production centre, the hot cell facilities, laboratories, decontamination facilities, etc., have also contributed significantly to the waste quantities. Necsa also accepted industrial and medical radioactive waste from smaller waste producers in the nuclear industry and the medical sector.

The bulk of Necsa's radioactive waste (intermediate-level waste, ILW, and high-level waste, HLW) is currently stored in various interim storage

20.2 Uranium conversion plant at Necsa [1].

facilities on the Necsa site (Fig. 20.3). These wastes are mostly contained in metal and concrete storage containers. The waste in containers varies widely in type (powders, filters, oil, etc.) and only ILW is encapsulated into a cement waste form. These wastes can be regarded as historical waste in that they were produced in the absence of a well-defined end-point (repository) and therefore in the absence of formal waste acceptance criteria.

The main generators of current/future SRAW in South Africa are Necsa, Koeberg power plant and the mining industry. The other generators such as the iThemba accelerator facilities, hospitals and industries are regarded as minor contributors. Koeberg generates low- and intermediate-level waste (LILW) and ILW that is sent to the national waste disposal site called Vaalputs, situated in the Northern Cape (Fig. 20.4). Koeberg also generates HLW. Currently, all the HLW (SF) is stored in the SF pool at Koeberg (Fig. 20.5). Dry storage of HLW (SF) at Vaalputs as an interim solution could be considered.

Current and future nuclear activities at Necsa will continue producing operational radioactive waste, albeit in a more controlled manner to comply with formally defined waste acceptance criteria [2]. Current activities at Necsa that generates waste are:

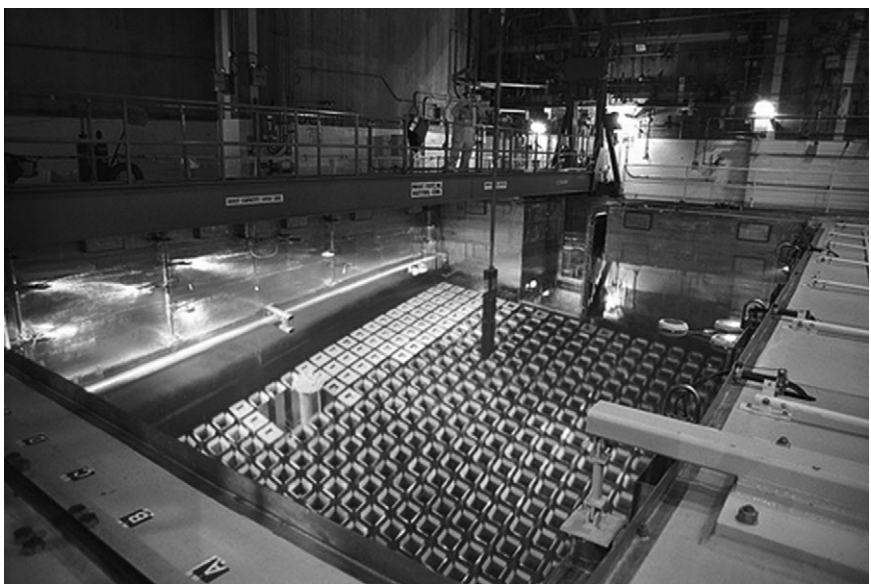
1. Nuclear fuel cycle
   - Uranium conversion (future)
   - Uranium enrichment (future)
   - Fuel fabrication (future)

(a)

(b)

20.3 (a, b) Main dry storage facility at Necsa site [1].

20.4 Vaalputs disposal site [1].



20.5 Storage of HLW at Koeberg [1].

- Fuel reprocessing (future)
- Decommissioning (current and future)
- SAFARI (current materials test reactor (MTR) in South Africa)

2. Supporting facilities
   - Laboratories
   - Research and development
   - Hot cells
   - Maintenance
3. Current operations
   - SAFARI 1 (research reactor)
   - Nuclear technology products (NTP – production of radionuclides)
   - Target plate manufacturing
4. External
   - Health care waste
   - Industrial waste
   - Spent sealed radioactive sources
5. Decommissioning waste
   - The research reactor utilized in the process for the generation of radioactive isotopes for industrial and medical applications
   - The liquid waste treatment facility producing sediment that is conditioned and classed as SRAW
   - Research laboratories
   - Fuel and target plate manufacturing centre
   - Decontamination facility
   - Various decommissioning projects.

The *Radioactive Waste Management Policy and Strategy* [3] for the Republic of South Africa was approved by cabinet in November 2005, in which certain structures are to be established:

- National Committee on Radioactive Waste Management (NCRWM),
- Radioactive Waste Management Institute, and
- Radioactive Waste Fund

On a strategic level, the National Radioactive Waste Management Policy and Strategy (NRWMPS) expresses the national commitment towards the management of RAW in order to ensure a coordinated and cooperative approach to RAW management and to provide a national strategy and framework for the development of future waste management plans. Legislation is currently being prepared to establish the National Radioactive Waste Management Agency (NRWMA) as an independent government-sponsored agency responsible for the disposal of all RAW on a national basis. This agency is expected to be in operation within the next three to five years.

20.2 Sources, classification and types of wastes

20.2.1 Sources of waste in South Africa

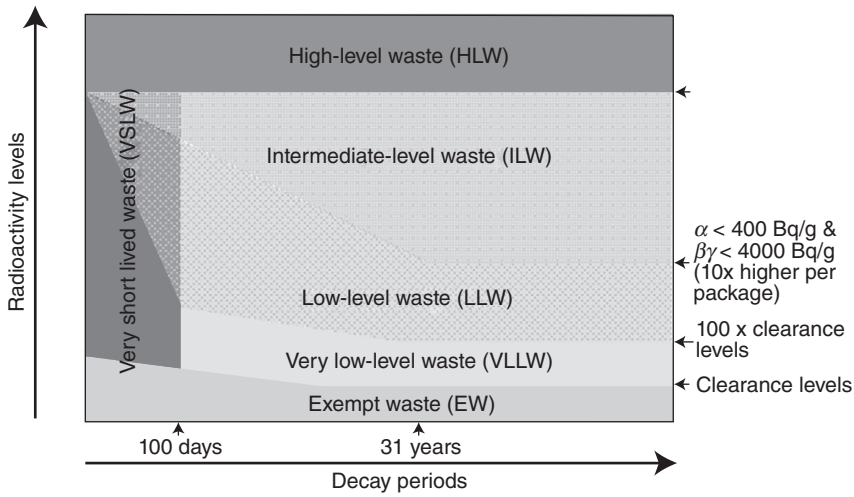
Hitorical waste at Necsa from the NFC programme includes waste from:

- the SAFARI-1 research reactor
- the development of a uranium conversion and enrichment capability
- establishment of research work on the NFC
- conduction of research with regard to the development of other types of nuclear reactors
- nuclear weapons programme.

20.2.2 Classification of RAW in South Africa

The RAW classification scheme as applicable to Necsa is generally in compliance with the scheme as published in the NRWMPS [3]. The proposed scheme for Necsa also makes provision for the latest IAEA international developments in RAW classification [4]. More waste classes are covered to address all the various waste streams of Necsa (e.g., special waste). The additional waste classes could be regarded as sub-classes of the main classes [3] which facilitate effective waste management and are complementary to the management methodology. The latest waste classification guidelines [4] are based on long-term safety aspects and do not specify criteria to distinguish between the classes. For the Necsa classification scheme, some criteria are maintained in view of the following:

- Although no medium depth (higher confinement) repositories currently exist in South Africa (still in the planning phase), waste needs to be classed in terms of general criteria for effective pre-disposal management. Waste characterized in terms of general criteria will be considered in the long-term safety assessments that are necessary for the authorization of such repositories. Taking a retrospective approach, the design of repositories will have to be suitable for waste that has been processed and is in compliance with specific long-term safety-related criteria.
- The long-term safety of the national near-surface repository for LILW at Vaalputs in the Northern Cape is demonstrated and is currently authorized in terms of specific criteria. The long-term safety assessment of Vaalputs needs to be reviewed in terms of specific criteria prior to the authorization of receipt of waste from different generators. This is necessary to evaluate the suitability of the disposal system at Vaalputs for specific waste streams and additional inventories.



20.6 Solid radioactive waste classification system.

Table 20.1 Waste classification scheme

Waste class at Necsa	National waste classification scheme [3]
1. HLW	1. HLW
2. ILW	2. LILW-LL
3. LLW	3. LILW-SL
4. VLLW	4. VLLW
5. EW	5. VLLW (exempt waste included in definition)
6. VSLW	6. Not included as waste class. Covered as waste treatment to reach exemption levels

The Necsa solid radioactive waste classification system is presented schematically in Fig. 20.6. The classification scheme also complies with the general classification scheme as indicated in Table 20.1.

20.2.3 Types of radioactive waste classes at Nesca

High-level waste (HLW)

High-level waste is heat-generating waste (typically above 2 kW/m^3 or waste that needs to be managed in terms of its heat-generating properties over long durations) with high long- and short-lived radionuclide concentrations which include fission products and actinides. SF pellets and element sections from post-irradiation testing of pressurized water reactor (PWR) fuel, waste from Mo production and SF from the SAFARI reactor are retained waste that is regarded as potential HLW. Potential HLW is retained in the

facilities of origin or stored in interim storage facilities in accordance with facility-specific nuclear authorizations. Potential HLW with proven heat-generation capacity of less than 2 kW/m^3 or waste that does not need to be managed in terms of its heat-generating properties may be considered for re-classification as ILW. Waste types with long-lived radionuclide concentration levels that would result in an inherent intrusion dose of more than 100 mSv/a, after an institutional control period of 300 years, shall be managed as HLW.

HLW that is removed from authorized containment systems shall be processed to ensure a solid waste form in a waste package that is suitable for handling, transport and storage for a period of 100 years. Disposal of HLW is limited to a high degree of containment and isolation from the biosphere over long time periods, which is obtainable by regulated deep geological disposal (hundreds of metres).

Intermediate-level waste (ILW)

ILW is waste which has limited heat generation capacity that need not be considered for its disposal or in its disposal option (typically below 2 kW/m^3) with intermediate short-lived and/or intermediate long-lived radionuclide concentrations. ILW consists mainly of irradiated uranium (uranium, actinides, other activation products and fission products) in smaller quantities, or cooled irradiated uranium or in the form of irradiated uranium contaminated waste as generated in the isotope production facilities and the operation of the SAFARI research reactor. Unirradiated uranium (from the NFC) could also fall into this waste class, especially when it occurs in higher concentrations. Waste management is aimed at preventing unirradiated uranium falling into this waste class. Long-lived sealed sources, for example Ra-226 or SHARS (spent high activity sealed radioactive sources) could also fall in this waste class. SHARS of shorter lived radionuclides may have such high activity levels that the intrusion dose after the institutional control period is in excess of 10 mSv/a as specified for near surface disposal.

The long-lived radionuclide half life ($T_{½}$ > 30.2 years) concentrations could on average typically be 4,000 Bq/g and 40,000 Bq/g for α and $\beta\,\gamma$ emitters, respectively. Criteria for long-lived radionuclide concentrations need to be justified for a specific repository. Criteria are justifiable in the case of a specific repository if inherent intrusion dose after the institutional control period is between 10 mSv/a and 100 mSv/a.

For irradiated uranium waste, containers are shielded to ensure surface dose rate levels <2 mSv/h. Unirradiated uranium waste could be pre-treated in unshielded containers. ILW that is removed from authorized containment systems shall be processed to ensure a solid waste form in a waste package

that is suitable for handling, transport and storage for a period of 50 years. Additional requirements may be prescribed for a specific ILW repository. Corporate pre-disposal management standards need to be specified to ensure good practice and waste packages that are compliant and verifiable in terms of the applied IAEA standards [5]. These generic standards ought to be used as reference standards for the evaluation of ILW disposal concepts and for the long-term safety of planned ILW repositories. Disposal of ILW needs a high degree of containment and isolation from the biosphere over a long period of time that is obtainable by intermediate disposal at depths of tens to hundreds of metres.

Low-level waste (LLW)

LLW is waste with low long-lived radionuclide concentrations and intermediate short-lived radionuclide concentrations. LLW consists of waste contaminated with unirradiated uranium from the NFC and waste contaminated with irradiated uranium in the form of activation and fission products from isotope production and the operation of the SAFARI research reactor and short-lived sealed sources with limited activity levels (sealed sources that would not exceed the inherent intrusion dose of 10 mSv/a after the institutional control period).

The long-lived radionuclide ($T_{½} > 30.2$ years) concentrations are limited to 400 Bq/g and 4,000 Bq/g for α and $\beta\,\gamma$ emitters, respectively. Factor of ten higher concentration levels are allowed per waste package. Deviation from the above long-lived radionuclide concentration criteria is justifiable in the case of a specific repository if the inherent intrusion dose is less than 10 mSv/a after the institutional control period. For LLW with higher concentrations of short-lived activation and fission products, containers are shielded to ensure surface dose rates of <2 mSv/h. LLW could be pre-treated in unshielded containers in case the surface dose is <2 mSv/h or if the surface dose rate will be <2 mSv/h after waste treatment (e.g., decay). LLW shall be processed to ensure a solid waste form and a waste package that is in compliance with the approved waste acceptance criteria (WAC) of the national near-surface repository of South Africa (Vaalputs) that is suitable for handling, transport and storage for a period of 10 years. LLW shall be disposed of at Vaalputs near-surface repository.

Very low-level waste (VLLW)

VLLW is waste with very low radionuclide concentrations. VLLW consists mainly of bulk quantities of waste due to the operation and decommissioning of nuclear facilities. VLLW has radionuclide concentration levels slightly above the levels specified for clearance of material with a limited

radiological hazard potential that justifies limited radiation protection provisions. Pre-treatment of bulk quantities of VLLW as LLW would be cost intensive and not justifiable. VLLW could contain non-radioactive materials that render the waste hazardous. VLLW also needs to be classed and managed in terms of all its non-radiological hazards.

Subject to specific authorization, VLLW may be disposed of in engineered landfill facilities or surface impoundments, general waste landfill facilities and hazardous chemical waste disposal facilities. Authorized re-use of material (e.g., recycling of concrete as aggregate or use for road construction) may also be considered as a management option. Specific criteria are derived for a specific facility or management option. VLLW could have radionuclide concentration levels of up to factor 100 above the clearance/exemption criteria for engineered landfill facilities. Longer lived radionuclides could be more limiting, depending on the site factors and design, due to the longer duration for which safety has to be demonstrated. Longer term institutional control arrangements may also be necessary for the VLLW disposal facilities. Mixing and consolidation of different VLLW or potential VLLW waste streams could be justified in order to lower radionuclide concentration levels or to obtain a more stable waste form, taking into consideration the physical and chemical compatibility of waste streams. Existing bulk waste collection systems (e.g., evaporation ponds) could also be considered for conversion and authorization as VLLW engineered disposal facilities.

Exempt waste (EW)

EW contains such low concentrations of radionuclides that it does not require radiation protection provisions. EW may be re-used, disposed of in general waste landfill facilities, disposed of in hazardous chemical disposal facilities or be recycled. EW needs to be cleared/exempted in terms of approved clearance/exemption criteria [6].

Very short-lived waste (VSLW)

VSLW contains short-lived radionuclides with a longer lived radionuclide content within approved clearance/exemption criteria. VSLW is treated by decay to a point where the short-lived radionuclides have also reached approved clearance/exemption criteria, whereafter it is managed as EW.

20.2.4 Current waste inventory

The total quantity of waste for the respective waste classes on the Necsa site is indicated in Table 20.2. It should be noted that of the total quantity

Table 20.2 Quantity of waste classes currently at Necsa

Radioactive waste class	Quantity of waste (m^3)
High-level waste	100
Intermediate-level waste	25
Low-level waste	16,000
Very low-level uranium containing bulk waste	27,000
Very short-lived waste	<100

of LLW (16,000 m^3), about 6,000 m^3 is 'compressible'. The compressible waste will undergo some volume reduction treatment processes, resulting in an approximately 60% reduction of this volume of waste. Further segregation of some of the Necsa historical waste (compressible and non-compressible) would be a difficult and costly exercise resulting in unnecessary additional radiological exposures of workers. For these reasons, the proposal is that the historical compressible and non-compressible waste inventory should not be further segregated but should remain in their respective waste categories unless alternative opportunities arise.

The LLW and ILW classes further consist of fissile and non-fissile material. Some of the material, especially the fissile categories, may contain sufficient quantities of uranium that may render recovery of this material worthwhile. For those waste streams, existing or new processes will be utilized or developed and only the resulting waste that cannot in any way be further reduced in volume or quantity will then be disposed of.

20.3 Radioactive waste (RAW) management strategies

20.3.1 Guiding principles

The following principles are applicable for the management of radioactive waste by Necsa [7]:

- Waste management is aimed at optimization of the processes from waste generation to waste disposal.
- The hierarchy for the selection of waste management options are as follows:
  - waste prevention and waste generation control
  - waste clearance
  - waste re-use and recycling
  - waste conditioning and storage
  - waste disposal.
- The principle of 'continuous improvement' shall apply to all waste management programmes and is addressed as a minimum requirement in

terms of Necsa's as low as reasonably acceptable (ALARA) programme and environmental management system.
- Final disposal is regarded as the ultimate step in the RAW management process, although a step-wise waste management approach is acceptable. Long-term storage of certain types of wastes (e.g., HLW, LLW and spent sources) may be regarded as one of the steps in the management process.
- The aim shall be to achieve a maximum degree of passive safety in storage and disposal.
- The establishment, operation, decommissioning and closure of waste-generating and disposal facilities shall be in accordance with all applicable regulatory requirements in force at the time.
- Radioactive waste management at Necsa shall cover the total life cycle of waste management, from generation to institutional control over closed radioactive waste disposal facilities.
- The transfer of waste among generators shall be considered provided all issues pertaining to ownership, liability and safety are addressed.

There are currently still some aspects with regard to waste management on a national as well as Necsa level that need to be resolved. In order to develop the Necsa Waste Management Methodology and the subsequent National Radioactive Waste Management Plan (NRWMP), certain assumptions are made as described in the following sections.

Disposal end-points

It is the responsibility of the NRWMA to provide final end-points for the respective waste classes. It is therefore assumed that an end-point for all waste categories will exist. The following assumptions are made at this point with regard to the Necsa RAW (the waste is classified in accordance with the Necsa waste classification scheme [8]):

- HLW: No end-point has as yet been decided upon on a national level. For the purpose of this document, disposal in a deep geological facility is assumed for this waste category. On-site storage is not suitable for the long-term storage of this waste class [3]. Interim storage off-site before final disposal will, however, form part of the waste management plan for this waste class.
- ILW (this may also include long-lived sealed radioactive sources such as Ra-226 as well as SHARS): Disposal in a greater confinement trench at the Vaalputs National Radioactive Waste Disposal Facility [3].
- Low-level solid waste (this includes short-lived sealed radioactive sources with limited activity): Disposal in a near-surface disposal facility. The Vaalputs National Radioactive Waste Disposal Facility is identified as the end-point for this waste class [3].

- Very low-level uranium containing bulk waste: Disposal on a mine slimes dam or, with special authorization, disposal in engineered landfill facilities or surface impoundments, general waste landfill facilities and hazardous chemical waste disposal facilities.
- Very short-lived waste: Exemption/clearance will be the final end-point for this material (sealed sources with half-lives <100 days) after being kept on site until sufficiently decayed.

Storage duration

All radioactive waste generated on the Necsa site is still stored there. However, on 7 May 2011, the first shipment of LLW (Fig. 20.7) was disposed of at Vaalputs, and as this operation will take place over many years, it is assumed that some of these waste classes will continue to be stored for at least another 10 years at the Necsa site. Conditioning of this waste should therefore allow for storage at Necsa for a period of at least 10 years. Conditioning of HLW should ensure compliance with long-term interim storage at the Necsa or Vaalputs site (50 years).

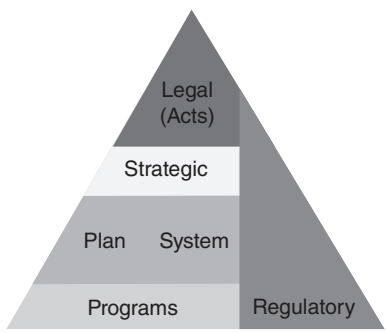
20.7 LLW from Necsa to transferred Vaalputs disposal site [1].

20.3.2 National radioactive waste management system

Radioactive waste management as interpreted by Necsa is structured as presented in Fig. 20.8. Radioactive waste management in South Africa is structured and implemented by including the applicable sections from the various National Acts, i.e National Environmental Management Act (No. 107 of 1998), The National Nuclear Regulator Act (No. 47 of 1999), the Nuclear Energy Act (No. 46 of 1999), the Hazardous Substances Act (No. 15 of 1973), the National Water Act (No. 36 of 1998), etc. On a strategic level, the NRWMPS expresses the national commitment towards the management of RAW in order to ensure a coordinated and cooperative approach to RAW management and to provide a national strategy and framework for the development of future waste management plans.

Site-specific waste management plans are then developed based on the directives and guidelines provided by the NRWMPS. The purpose is to create an optimized and sustainable plan that provides for acceptable waste stream-specific pre-disposal management prescriptions for the identified waste end-points. The waste management plan is supported by the waste management system elements aimed at ensuring and demonstrating that waste management practices comply with requirements. The system renders the support structure for the implementation of the site-specific waste management plans. It provides for pre-disposal management standards and integrates the relevant legal, regulatory and strategic management requirements that will eventually lead to the National Nuclear Regulator (NNR) approval of the system. Elements of the System include: site waste management principles, site waste management responsibilities, quality assurance and site waste management processes and pre-disposal standards.

Facility-specific waste management programmes (FSWMP) are developed from the waste management plan. The purpose of this is to ensure and demonstrate compliant and consistent waste management practices at each facility. These programmes are developed by integration of the relevant plan, system, operational and regulatory requirements.



20.8 Waste management framework.

The general radioactive waste management process is demonstrated in Fig. 20.9, which shows the typical interactions between waste generators, pre-disposal and disposal operators, as well as the demarcations existing between them. It should be noted, however, that Fig. 20.9 presents the process for general waste streams. In the case of special waste streams, the waste generators are responsible for some or all of the pre-disposal steps.

20.3.3 Waste management processes

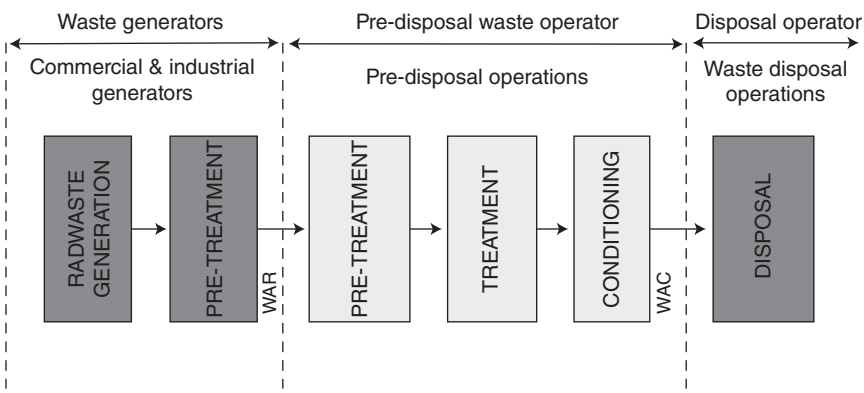
Waste prevention and generation control

The over-arching principle in radioactive waste management is to prevent the generation of waste and to keep waste volumes and activities to the minimum practicable. This is obtained through appropriate design measures and operating practices [9]. If waste generation can be prevented, there will be no need for further management. Necsa have implemented the following basic principles with regard to waste management.

Pre-treatment

Pre-treatment constitutes all the operations prior to waste treatment, such as collection, segregation, chemical adjustment and decontamination and is performed at the stage of waste generation. Pre-treatment may result in a reduction in the amount of waste requiring further processing and disposal.

Pre-treatment activities should be conducted in a manner that minimizes the volume of primary and secondary RAW requiring treatment and minimizes the volume of stored or disposed waste. Management options such as recycle, re-use and clearance, should be implemented as far as reasonably practical.



20.9 Radioactive waste management process.

The first operation in pre-treatment is to collect waste, then segregate it on the basis of the waste stream's physical state (liquid, gaseous, solid), activity concentration and total radioactivity. RAW is segregated to avoid mixing waste streams. Short-lived radionuclides should not be mixed with waste containing long-lived nuclides. The segregation strategy should also assess whether the waste can be cleared, either directly or after some period of decay, or recycled.

Waste streams should preferably not be mixed due to different downstream processing methodologies. There are, however, some instances when mixing of streams is an acceptable practice, for example:

- waste streams requiring the same downstream processing or disposal route,
- waste volume from one or more of the individual sources too small to justify investment in specialized processing.

Treatment

Treatment consists of operations intended to provide safety or economic benefit by changing the characteristics of the waste. Three basic treatment objectives in waste treatment are:

- Volume reduction (incineration of combustible waste, compaction of solid waste, disassembly of bulky waste components or equipment).
- Removal of radionuclides from the waste (e.g., decontamination, melting of contaminated metal components, evaporation of liquid waste streams and filtration of gaseous waste streams). Decontamination can result in volume reduction by clearing waste or changing waste class to waste with end-point.
- Change of physical or chemical composition of the waste (e.g., solidification of sediment to enable disposal).

Conditioning

Conditioning consists of those operations that produce a waste package suitable for handling, transport, storage or disposal. Prior to conditioning RAW for storage or disposal, the pre-disposal management waste acceptance requirements (WAR) and the disposal facility WAC have to be considered to ensure compliance with the storage facility or disposal site requirements, respectively. Where final disposal criteria do not yet exist, disposal criteria assumptions will be defined and incorporated into processing methodologies.

It should be noted that for some waste streams, treatment actions render a waste package that already conforms to the criteria for disposal and that

no further conditioning is required. After conditioning, the final characterization will take place in order to ensure that the waste package conforms to the WAC of the disposal facility.

Responsibilities

The responsibilities of the 'generators and operators' entail the following [3]:

- The technical, financial and administrative management of such wastes within the national regulatory framework and within any applicable co-operative governance arrangements.
- Development and ongoing review of site/industry-specific waste management plans which are to be based on the requirements stipulated in the national radioactive waste management policy and strategy.
- Implementation of waste management plans by the establishment of appropriate waste management and facilities processes and the development of site/industry-specific waste management systems.
- Site/industry waste management in accordance with waste management systems to reflect sustainable development and principles such as continual improvement and best available technology not entailing excessive cost (BATNEEC) and other elements of the national strategy.

At Necsa, the responsibility for the management of solid RAW is documented [10]. This document highlights the responsibilities of the waste generators, the pre-disposal waste operator (NLM), waste disposal operator (National Radioactive Waste Disposal Institute) and the Safety Health and Environmental Quality Department (SHEQ).

The responsibility for the development and maintenance of the NRWMP lies with the Nuclear Liabilities Management (NLM) department of the Nuclear Services Division of Necsa. NLM is also responsible for the submission of the plan to the National Committee on Radioactive Waste Management (NCRWM) [3].

The establishment of the NRWMA and the main responsibility of the NRWMA is the final disposal of waste on a national basis at the Vaalputs National Radioactive Waste Disposal Facility, to ensure correct siting and design and to construct and operate new RAW disposal facilities for other waste categories [3].

One of the functions of the NRWMA is to assist generators of small quantities of radioactive waste with the management of such waste as well as the management of ownerless waste (e.g., orphan sources) on behalf of the government. These responsibilities are currently being expedited by Necsa (NLM) on behalf of the government. The management of such waste streams are therefore included in the NRWMP.

20.4 Waste acceptance criteria (WAC)

WAC have been established for the LILW disposal facility at Vaalputs. The WAC are important in the RAW management chain as they determine the treatment and conditioning processes and requirements for waste streams which will form the final waste package for disposal. The WAC for some of the waste, especially in situations where end-points have not yet been decided upon, have not been finalized. Since the WAC dictate the waste treatment and conditioning processes, generic WAC are therefore assumed.

20.4.1 General requirements

1. Waste will be managed in accordance with the Necsa waste management system [10].
2. Each waste generator shall prepare a facility-specific waste management programme which must be accepted by pre-disposal operations (PDO) [3].
3. All waste streams shall be listed and described in the facility waste management programme. New waste streams shall only be accepted after being included in this programme and accepted by PDO.
4. The collection, segregation and pre-treatment of waste at the generator shall take cognisance of the Necsa Waste Management Plan. This entails the alignment of the respective waste streams generated with the waste management processes and disposal end-point envisaged for each respective waste stream.

20.4.2 General waste accepted requirements

1. Only LLW and ILW radioactive waste that has been conditioned into a solid shall be accepted at Vaalputs disposal repository. This includes solidified waste streams such as sediments, resins and sludge.
2. Radioactive waste shall be safely managed in a regulated manner, compatible with internationally and nationally agreed principles and standards.
3. The waste generator shall demonstrate that the waste packages comply with the Necsa quality standards for the manufacture of containers and the filling and storage of those waste packages [11].
4. PDO will have the right to perform audits on the waste generators' waste management system and the implementation thereof.

20.4.3 Prohibited waste

1. Liquids and gaseous substances will not be accepted.
2. Waste packages must not contain free-standing liquid.

3. No un-stabilized explosive or untreated pyrophoric material will be accepted.
4. No compacted, solidified or conditioned waste shall be accepted at solid waste operations (SWO) unless such waste has been subject to a qualified and NLM-approved conditioning process.

20.4.4 Radiological characterization requirements

1. The applicable radionuclides to be expected in a waste drum shall be provided for each drum.
2. Waste drums shall be radiologically characterized, reporting the activity of all nuclides present in the waste.
3. Radiological characterization of waste drums can be carried out by non-destructive assaying (NDA) such as a drum scanner (Fig. 20.10) or radiochemical analysis of representative sample/s taken from the drum.
4. If use of statistical sampling is considered (e.g., limited number of drums analysed from a whole population), the suggested method shall be documented, justified and presented to PDO for approval, before any such sampling and analysis would be accepted.
5. NLM has a NNR-authorized radiological characterization (NDA) method for waste drums containing gamma-emitting nuclides and waste density of less than 1 g/cc in 100–210 L plastic or metal drums.
6. Waste from generators that could be characterized by this authorized method can be accepted without the need to provide evidence of radiological characterization.
7. The characterization of any waste drum containing a mixture of gamma- and non-gamma-emitting nuclides, or pure non-gamma-emitting nuclides, or density more than 1 g/cc lies with the waste.
8. When analyses are to be based on analysis of a representative sample taken from the waste, the analysis shall be performed by an accredited laboratory and method, and samples taken according to an approved sampling plan. This plan shall provide evidence that the prescribed sampling method ensures representative sampling. Evidence of at least the following shall be provided with each waste drum:
   - sample and waste mass
   - traceability to calibrated scales used
   - activity calculation sheet
   - laboratory analysis report
   - reference to approved characterization procedure.
9. Evidence of the approved characterization method shall be provided to PDO, and where applicable a copy of the approved sampling plan.

20.10 Non-destructive assaying at Necsa [1].

10. When none of the methods described is possible, generators could still apply to PDO for acceptance of the waste by submitting a motivation for acceptance of a best estimate of the activity in the waste package based on knowledge of the waste producing process. A formal request shall be provided where at least the following shall be provided in a report:
    - unique identifiers of waste drums (numbers)
    - detailed description of waste and container
    - quantities, number of drums and weight
    - reasons why characterization (sampling/analysis, etc.) is not possible
    - detailed description of the assumptions made and justifications for the expected nuclide-specific activity in each drum
    - nuclide-specify activity estimations for each drum.

20.4.5 Chemical characterization requirements

1. Details shall be provided on the chemical contaminants that could be present in the waste. This could typically be based on the chemicals used in the processes from which the waste originates.
2. Pre-treatment of waste by generators should take place only after consultation by PDO or if approved in the facility-specific waste management programme. If any pre-treatment is performed by the waste generator, details on this process, material or chemical added, etc., shall be provided. Quantities, mass and ratios of pre-treatment material or chemicals where applicable shall be provided.

20.4.6 Radiological acceptance requirements

The following criticality limitations are applicable to the WAC [12]:

- 100 L to 160 L drum: total ^{235}U mass <200 g per drum
- 210 L drums containing compressed drums: total ^{235}U mass <500 g per drum
- 210 L drums containing any other waste: total ^{235}U mass <250 g per drum.

More specific requirements are stated in Section 20.4.9.

20.4.7 Waste form requirements

1. The waste form shall be passively safe without the possibility of internal corrosion of waste container caused by the waste form or the possibility of volume increase of the waste form due to formation of corrosion products.
2. Pre-treatment, treatment and immobilization actions should be aimed at providing passively safe waste forms.
3. The waste form does not contain or have the potential to generate hazardous or corrosive materials unless it is demonstrated that the encapsulating or immobilizing matrix of the waste form makes them passively safe.
4. If waste drums contain uranium metal, the following limitations are applicable:
   - items shall be dry (no water, oil or grease)
   - volume per item not less than 5 cm^3.
   - surfaces of all items to be smooth
   - minimum thickness 10 mm.

20.4.8 Package requirements

The maximum permitted surface dose rate for each package for storage (drums, concrete containers, ISO containers and ingots) shall not exceed 1 mSv/h at 0.5 m and 2 mSv/h for contact dose rate. The maximum permitted removable surface contamination on waste containers shall not exceed 0.04 Bq/cm^2 for α contamination and 0.4 Bq/cm^2 for β contamination.

The following information shall be clearly indicated on the drum or on a label affixed to the drum:

- unique container number traceable to the information/documentation accompanying the package
- gross mass
- date when filled
- type of waste
- maximum contact dose rate
- dose rate at 0.5 m.

If containers are used with any other specific additional liner/coating to protect the surface of the drum, details describing these liner/coatings shall be provided in the facility-specific waste management programme.

The mass of any waste package shall not exceed the manufacturer's loading limit as specified for each drum type/design. Should the waste generator have to exceed this limit, prior approval from the manager PDO is required. A report justifying the use, giving evidence of compliance to IAEA transport regulations [13] and metal drums specification [14] shall be prepared and submitted to PDO. An example of current waste packaging is shown in Fig. 20.11.

The following waste containers will not be accepted:

- waste containers coated in an attempt to cover cracks or corroded surfaces
- double packed drums (waste placed inside a drum and again inside another drum).

20.4.9 Specific requirements for unconditional waste

Segregation of waste

Unconditioned waste shall be segregated as follows:

1. compressible waste
2. non-compressible waste
3. uranium contaminated steel and metal waste (excluding lead), which shall be segregated in the following sub-groups:
   - mild steel
   - galvanized steel

20.11 Current ILW waste package used in South Africa for disposal [1].

- stainless steel
- aluminium and
- copper.

Specific packaging requirements

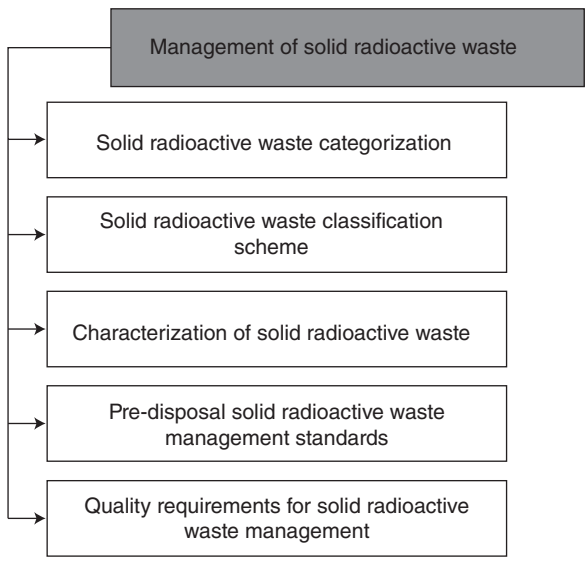
1. Compressible waste shall comply with the following criteria:
   - waste must be packaged inside a metal drum
   - the drum shall be minimum 100 L and maximum 160 L drum
   - the drum wall thickness shall not be less than 0.6 mm
   - the drum's diameter shall not be less than 25 cm
   - less than 20% elastic material in waste
   - the mass of the drum contents shall not exceed the manufacturer's safe working load.
2. Non-compressible waste must be packaged inside a metal drum and shall comply with the following criteria:
   - the drum shall be a 210 L drum
   - the drum wall thickness shall be not less than 0.8 mm
   - the mass of the drum contents shall not exceed the manufacturer's safe working load.

3. Uranium contaminated steel or metal waste:
   - this waste shall be contained in 210 L drums or ISO containers (shipping containers)
   - the mass of the drum contents shall not exceed the manufacturer's safe working load.

20.5 Necsa solid waste management system

The Necsa solid waste management system is aimed at providing principles, guidelines and standards that are aligned with sound waste management practices. The system is also aimed at managing the interfaces that exist in terms of the waste management steps and the Necsa organizational structure.

The principles of waste prevention and waste minimization are paramount in the overall Necsa waste management system. These principles are entrenched in every step of the waste management process, from the point of operations in the various Necsa facilities to the decisions on the options for the management of different waste categories. Although not being taken into account in the past, it is also important to consider waste generation control and minimization during the design phase of future facilities at Necsa. The design of buildings and plant should be such that the minimum amount of waste is generated by the planned activities. This integrated system is shown in Fig. 20.12.



20.12 Integrated solid waste management system.

20.5.1 Solid radioactive waste categorization

A system is provided according to which radioactive waste on the Necsa site shall be categorized to enable the identification of waste for subsequent waste management processes. The scheme for the categorization of waste as well as the principles for categorization of radioactive waste is addressed in this systems document.

20.5.2 Solid radioactive waste classification scheme

The process involves the implementation of the national radioactive waste classification scheme in the Necsa context in support of solid radioactive waste management on the Necsa site at Pelindaba. The system is aimed at presenting the classification scheme as it applies to Necsa and providing principles and guidance on waste classification in terms of the approved classification scheme [3] and latest international developments in this regard [15]. The system is further aimed at applying the waste classification criteria to typical waste streams at Necsa.

20.5.3 Characterization of solid radioactive waste

This provides for a system to characterize waste in terms of its radiological, physical, biological and chemical properties. It determines the needs for further adjustment, treatment, conditioning, or its suitability for further handling, processing, storage or disposal.

20.5.4 Pre-disposal solid radioactive waste management standards

The standards prescribe the general pre-disposal waste management considerations, guidelines and standards for Necsa and also provide a framework for the development of specific pre-disposal waste management standards for inclusion in the 'facility specific solid radioactive waste management programmes'.

20.5.5 Quality requirements for solid radioactive waste management

The arrangements for compliance assurance during all waste management process steps ensure proper implementation of the waste management system. This quality management programme will provide the guidelines for ensuring that

- Facility-specific waste management programmes are developed in accordance with the Necsa waste management plan and system requirements
- The facility-specific waste management programmes are properly implemented
- Proper quality control is executed during all waste management processes (waste generators and waste operators).

20.6 Necsa radioactive waste management plan development

The national radioactive waste management policy and strategy document [3] prescribes the use of a balanced and systematic way of evaluating respective waste management options using a multi-attribute analysis approach such as the BATNEEC process. It was, however, decided to utilize the more recent best practical environmental option (BPEO) and best practicable means (BPM) multi-attribute analysis processes. The BPEO process will be utilized for the selection of the management options whilst the BPM process will be followed for the refinement of design and operational conditions.

BPEO studies are particularly relevant to strategic decision making, involving choices between alternative management options. The fundamental comparison relates to the performance of environmental options, but the process should provide a holistic appraisal, which includes continual improvement, of factors associated with the practicability of implementing strategic alternatives. BPM relates to optimization of the selected option from the perspective of radiological protection, and is concerned with the detailed refinement of design and operation conditions.

The evaluation criteria used for the selection of the BPEO and BPM are as follows:

1. Cost effectiveness
   - life cycle cost of waste.
2 Operational feasibility
   - existing or new technology
   - international best practice
   - regulatory constraints and challenges
   - ease of operation.
3. Environmental and social acceptability
   - public safety impact
   - perceived risk and social acceptability
   - environmental impact
   - continual improvement potential.

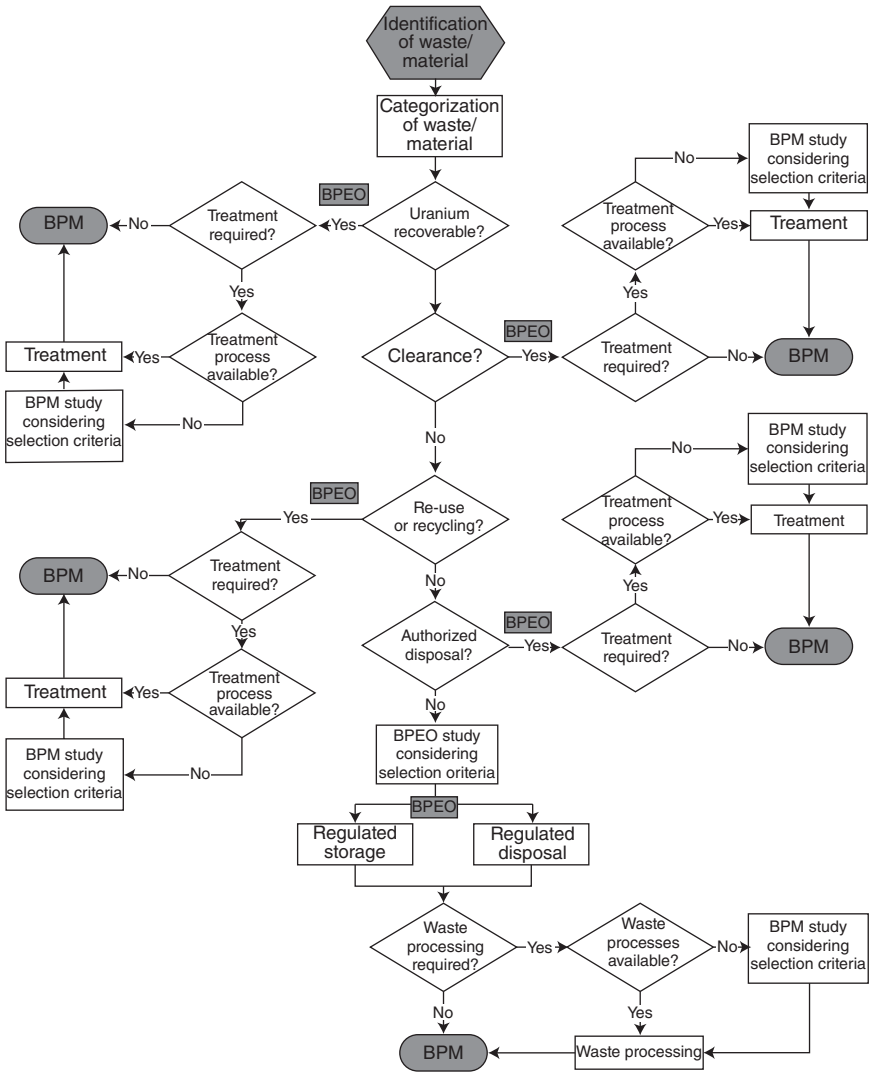
4. Safety
   - worker safety impact
   - public safety impact
   - accident risk
   - safety impact reduction potential.

The aim of a BPEO study is to ensure that the reasoning behind a strategic decision, involving technical, scientific and more qualitative judgments (including their consistency with the overriding principles of precautionary action and sustainable development), is made visible.

The process to determine the BPEO for each waste or material category is presented schematically in Fig. 20.13. In instances where management options for certain waste categories exist and those processes are authorized or in the process of authorization, then that will be regarded as the preferred option (BPEO) for that waste category. It should be noted, however, that the authorization process includes evaluation of options and justification of selected actions.

This process entails the following in sequential order:

1. Identification of the waste or material. This includes radioactive waste or redundant radioactively contaminated materials/equipment for which no further need is foreseen as identified by the generator or waste operator.
2. Categorization/grouping of waste/material according to its possible management option. The categorization allows for grouping together of waste streams or material with the same attributes and that will be managed in the same manner.
3. Following the flow diagram for each waste/material category to find the BPEO and associated BPM.
   - Recovery of uranium (source material). Some radioactive waste at Necsa contains uranium that could be recovered for re-use. Due to the high value associated with uranium, especially uranium in its enriched form, it is worthwhile to identify radioactive waste streams with recoverable quantities of uranium. The recovery route will be regarded as the BPEO for that respective waste stream. Different option evaluations will be performed in order to find the BPM for each waste stream in this waste category where recovery processes have to be developed.
   - Waste/material clearable. Material or waste that conforms to the criteria for clearance will not be further treated but will be processed to demonstrate compliance with clearance criteria. The clearance route will be regarded as the BPEO for that category. The 'clearance' of radioactive material allows for the release of the material from nuclear regulatory control in terms of the compliance with clearance



20.13 Process to determine BPEO for applicable waste management options.

levels. If the waste/material is clearable with further treatment, then it will be treated as such. If a treatment process exists, then treatment will be performed. If not, an exercise will be performed to find the BPM for treatment of that category as described previously. Existing treatment processes include chemical decontamination and smelting.

- Waste/material acceptable for re-use or recycling. This step allows for the release of material that does not conform to clearance criteria

but conforms to the criteria for re-use or recycling. If treatment is required to enable release of the material, the same procedure will be followed as described previously to determine the BPM.

- Waste/material acceptable for authorized storage.
- Waste/material acceptable for authorized disposal. This step allows for the release of material that does not conform to clearance criteria or for re-use or recycling but does conform to the criteria for authorized disposal. If treatment is required to enable authorized disposal of the material, the same procedure will be followed as described previously to determine the BPM.
- BPEO waste management option study (regulated storage or regulated disposal). Waste that does not conform to any of the above categories requires an option study to find the BPEO for that waste category. The option study will determine whether the BPEO for a specific waste category will be regulated disposal or regulated storage. Once the BPEO for that waste category has been selected, the waste will be treated in accordance with existing processes or, if such processes do not exist, an option study will be performed to select the BPM.

  There are three options when treating radioactive waste, namely volume reduction, removal of radionuclides from the waste and the change of the physical form and/or chemical composition. The nature and composition of the respective waste categories allow in most instances a decision to be taken on the BPM for that waste category. For example, in the case of the compressible waste type, the BPEO would be volume reduction. Different methodologies can, however, be applied in order to reduce the volume of the waste such as compaction, incineration and segregation. Option studies are subsequently performed in order to determine which of these methodologies would be the preferred option or for the respective waste category.

4. When the BPM evaluation process indicates the establishment of new facilities or processes or the significant modification of existing facilities or processes, then external regulatory approval is required. This will mainly entail the Environmental Impact Assessment (EIA) process as prescribed by the Environmental Management Act, No. 199 in order to obtain a Record of Decision (RoD) as well as the NNR's nuclear authorization process.

20.7 Development of the Necsa radioactive waste management plan (NRWMP)

The Necsa waste management plan methodology as described in this document provides the basis for the development of the NRWMP. The NRWMP

in turn will provide the general approach for the management of RAW on the Necsa site. It forms the basis of the complete Necsa RAW management process. Generators of RAW and the RAW management department together with the SHEQ department will align themselves in order to give expression to the contents of this plan. The actions to be followed after the evaluation of the methodology described in this document by the National Committee on Radioactive Waste Management and the subsequent approval by the Minister (DME) includes the following:

- Development of the NRWMP. It was decided to develop the NRWMP in two steps. The first step will address the Necsa historical RAW while the second step will address the Necsa current and future RAW streams. The two steps will be submitted to the national committee separately.
- Facility-specific radioactive waste management programmes. Developing of facility-specific radioactive waste management programmes by each radioactive waste generator. These programmes should take into account the system requirements such as the principles of waste prevention and waste minimization and should allow for the pre-treatment of the waste in order to conform to the Necsa waste management department's waste acceptance requirements.
- Full implementation of the Necsa radioactive waste management system.

The NRWMP finally aims to provide an overview of the RAW management processes at Necsa in an open, transparent way. It will ensure that all RAW generated during past, present and future operations will be dealt with in a responsible manner that will not present an undue burden on future generations and the environment.

20.8 Decommissioning strategies and planning

Similar to nuclear programmes worldwide, the uranium conversion and enrichment research and production projects in South Africa were terminated in the early 1990s. During this time, the decommissioning strategy was aimed at returning the Necsa site to greenfield. To achieve this, the final disposal of waste, demolition of all buildings and the remediation of a site to conditions prior to any development was considered. Currently, demolition is no longer considered as a final decommissioning phase and, as the demand for nuclear facilities increases (nuclear renaissance), redevelopment and re-use (R/R) after decommissioning are currently envisaged for the buildings on the Necsa site. This will ensure a holistic approach based on current and projected future redevelopment demands. The new redevelopment and re-use plan aims to allocate previously licensed buildings to similar or the same nuclear projects as housed originally in the specific building, thus meeting most of the design requirements. There are various

buildings on the Necsa site that are currently in a decommissioning or a care and maintenance phase that will now be evaluated to ensure the optimization of decommissioning costs and waste minimization. The possible reutilization of process equipment could prevent unnecessary generation of waste and the implementation of additional radiological protective measures resulting in decommissioning costs.

Currently, conceptual decommissioning plans exist for most nuclear facilities and these plans will be explored to include possible redevelopment options. Emphasis shall be on the preservation of buildings and infrastructure, to keep them structurally sound and operable. For example, the decommissioning, decontamination and possible reutilisation of the uranium conversion facility at Necsa (Fig. 20.14) could have a major influence on the new Necsa nuclear fuel cycle initiative's business strategy and plan and waste management. Decommissioning projects aim at waste minimization by ensuring effective equipment and technology are used and proper segregation of waste is applied.

20.9 Future trends

20.9.1 Nuclear reactor programme

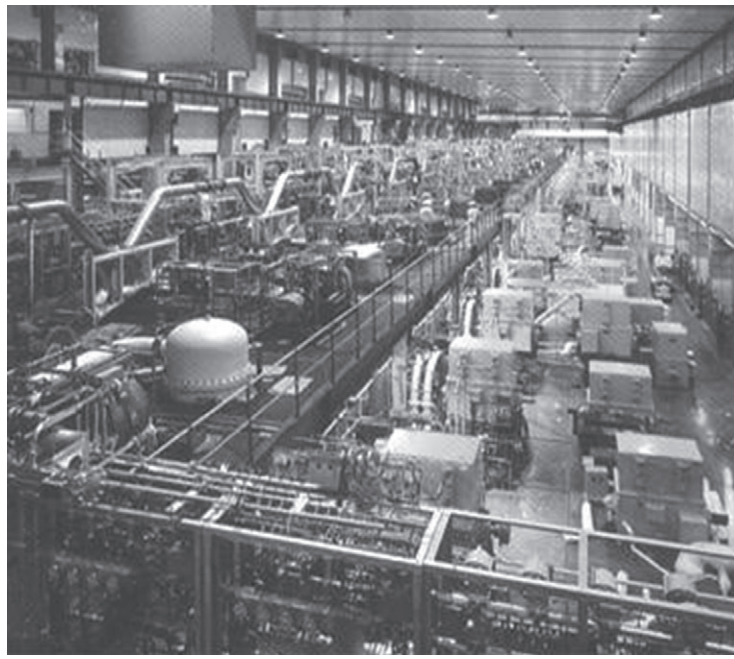
Currently, South Africa has two PWRs situated at Koeberg (Fig. 20.15), with a combined capacity of 1,842 MWe (6% of the country's electricity) [16].

Its first commercial nuclear power reactor began operating in 1984. As is made clear in its draft nuclear policy document [17], the government's commitment to the future of nuclear energy is strong. The state utility Eskom, which operates the Koeberg plant, has invited bids for a new nuclear power station. The new programme would start with up to 9,600 MW of PWR capacity by 2030, with the first unit commissioned in 2023. The environmental assessment process is under way, considering five sites, and the selection of technology will follow.

South Africa presently has a number of dedicated nuclear sites in reserve, and there are plans for the reservation of further nuclear sites. The existing Koeberg NPP could accommodate several additional reactors, but the intention is to establish a strategic reserve of nuclear sites.

20.9.2 Nuclear fuel cycle activities

During the time when economic sanctions were in force against South Africa, many nuclear fuel cycle activities were developed indigenously. Uranium production has generally been a by-product of gold or copper mining but, with the increased demand and prices today, further exploration is in progress. Originally, fuel for Koeberg was imported but, because of



(a)

(b)

20.14 (a, b) Decontamination of historical conversion plant to re-use facility [1].

20.15 Two PWRs situated at Koeberg [1].

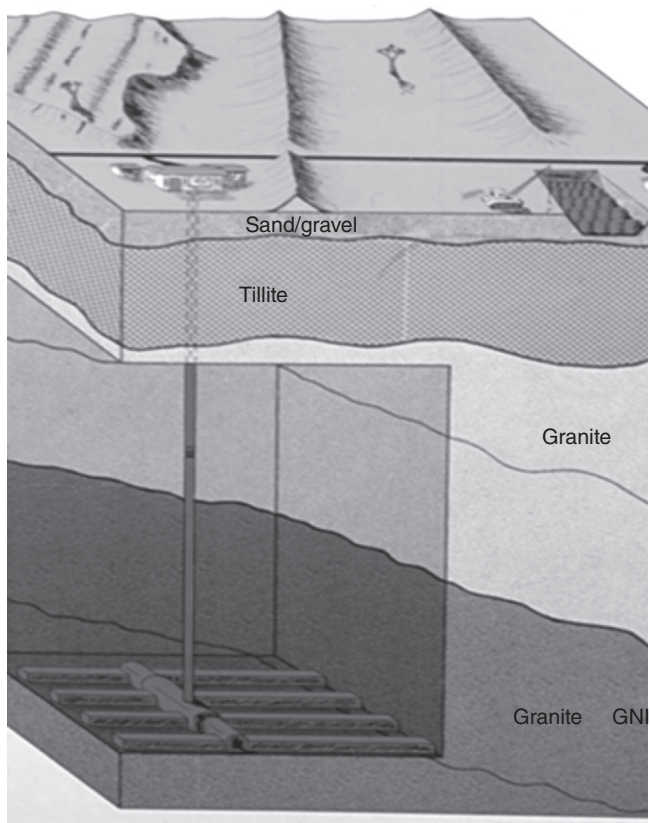
sanctions, the Atomic Energy Corporation set up conversion, enrichment and fuel manufacturing services for Koeberg. Enrichment was done at Pelindaba. For research reactor and military use, 45% enriched uranium was produced and for Koeberg low enriched material. Operations were halted in 1990 and 1995, respectively.

The new South African nuclear policy advocates re-development of the country's nuclear capabilities. It potentially allows for the country to implement conversion and enrichment facilities in order to gain more benefit from its uranium reserves. The ambitious programme goes still further and spans the full nuclear fuel cycle, to include fuel fabrication, reprocessing and recycling. The explicit policy goal is *'attainment of global leadership and self-sufficiency in the nuclear energy sector in the long term'* [17]. An investigation commissioned by the Department of Minerals and Energy concluded that it would not be advisable to exclude the reprocessing, conditioning and recycling of used fuel. Both national and foreign reprocessing options are conceivable and the government has requested that these options be investigated.

An *integrated* waste management strategy must take into account ***all*** of the radioactive wastes from ***all*** nuclear activities – in the past (legacy wastes), currently (mining, power production, medicine, industry and research) and in the future (decommissioning and, potentially, enrichment, fuel fabrication and reprocessing). This implies that, despite its present modest nuclear programme, South Africa must address a range of waste

management issues as wide as that in the most developed nuclear countries of the world. New issues will arise if fuel cycle activities are expanded. For example, experience in the UK and other countries has shown that, if reprocessing is undertaken, then waste streams become significantly more diverse and new questions, such as whether surplus plutonium is a resource or a waste, must be addressed [18]. This emphasizes the need for a comprehensive and integrated waste management strategy and operational programme. In the present report, however, attention is focused on the management of the wastes from power production and, in particular, on the SF and HLW.

Regardless of any HLW management strategy chosen in the future, a deep geological repository is needed, as long-term storage of SNF and HLW is not considered attractive but recognized as an interim option. Suitable high isolation environments are available in South Africa to host a deep geological repository. However, the development of a deep geological



20.16 Concept for a HLW repository at Vaalputs (1990s).

repository is a multidisciplinary process, by nature involving legal, technical, safety, economic, but also societal requirements/constraints. For the selection of a site for long-term management of SNF and HLW, public participation will be included.

The following has been completed as part of the process to establish a deep geological disposal repository:

- The potential of the current Vaalputs site to be used as a deep geological disposal site was investigated by Necsa during the early 1990s. The initial schematic concept can be seen in Fig. 20.16.
- Eskom completed feasibility studies with regard to geological disposal site selection, repository design, R&D requirements, interim storage, encapsulation plant, SNF transportation in 2007.
- Eskom 'Technical SF Management Plan' based on direct disposal (for costing and planning purposes).

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21

Republic of Korea: experience of radioactive waste (RAW) management and contaminated site clean-up

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Abstract: Republic of Korea currently operates 21 nuclear units providing one-third of the nation's electricity. Low and intermediate level radioactive materials emanating from these plants, medical facilities, research reactors, and industry need to be safely stored and managed. Disposal of spent nuclear fuel is also an important national issue. This chapter reviews the current state of affairs in Korea and examines the national policy, strategy, and direction for managing spent fuel and radioactive waste (RAW) materials. Decontamination of waste materials is also discussed.

Key words: Republic of Korea, radioactive waste (RAW), spent nuclear fuel (SNF) storage, disposal, decommissioning, decontamination.

21.1 Introduction

The twenty-first century's grand challenges are aptly characterized by energy, environment, and economy – the so-called tri-lemma of sustainability. These three Es are intricately interconnected, and balancing them is necessary for a healthy society. Many of this century's issues are global in nature, such as global warming that cuts across national boundaries and requires global cooperation in energy, environment, and economy to solve them. We are all in the same boat and must work together to meet these formidable challenges.

According to the International Energy Outlook 2011 reference scenario, the world's energy consumption is expected to grow by 53% between 2008 and 2035. Global electricity generation will grow from 19.1 trillion kWh in 2008 to 35.2 trillion kWh in 2035, an increase of 84%. Likewise, nuclear generation is expected to increase from 2.6 trillion kWh in 2008 to 4.9 trillion kWh in 2035. As for Korea, energy is particularly crucial for its national growth planning, as Korea has virtually no natural resources.

21.1.1 The energy situation in Korea

The energy situation in Korea is worse than in many countries, as Korea has no viable natural energy sources and must import primary energy. In

2011, Korea imported approximately 97% of its primary energy. South Korea is the world's No. 5 crude oil buyer and No. 2 liquefied natural gas importer and has boosted spending to acquire assets and develop oil and gas reserves, with a heavy focus so far on the Middle East and the Arctic. As a result, Korea is currently the ninth largest emitter of greenhouse gases in the world. Korea's greenhouse gas emission rates are increasing at the fastest rate (2.8%) in the world.

An important agenda in Korea's energy development plan is to promote nuclear power as a strategic response in the post-fossil fuel era and as a pillar of energy security and independence. Korea mapped out its long-term energy development plan based on the 3Es – energy security, economic efficiency and environmental protection. Korea hopes to reach its long-term energy goals by

- improving energy efficiency and reducing energy consumption,
- promoting clean energy including nuclear and renewable energy to reduce dependence on fossil fuels,
- boosting the green energy industry, and
- making energy sources accessible and affordable to low-income households.

Korea's total installed electricity generation capacity, standing at 72,491 MWe as of 2008, is projected to grow to 95,115 MWe by 2020 and further to 105,195 MWe by 2030. According to the Carbon Dioxide Information Analysis Center (CDIAC), Korea is the ninth highest country in carbon dioxide emissions in the period 1950–2005. USA (25%), China (10%) and Russia (8%) are the top countries in carbon dioxide emission in 1950–2005.

The Korean government is focusing its efforts on nuclear power as part of a national strategy to reduce greenhouse gas emissions and to achieve low carbon sustainable growth, Korea aspiring to become a green power country with low carbon, green growth. The national vision is to become the world's seventh largest green power by 2020 and the fifth largest green power by 2050.

21.1.2 Nuclear power in Korea

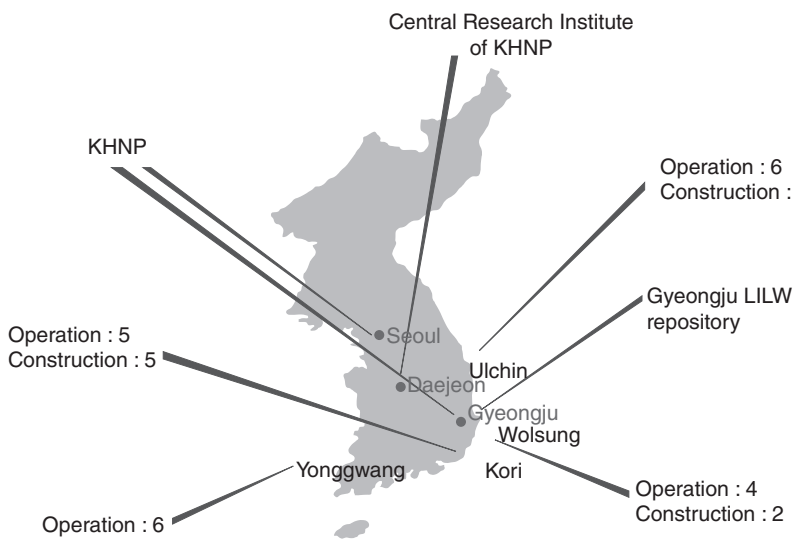
Korea's nuclear development has been robust and steady. The data shows an unplanned shutdown rate of 0.3 trips/reactor/yr and capability loss of 0.36% in 2009, the best record in the world. Its long-term energy plan entails increasing the nuclear installed capacity to 41% and nuclear generation to 59% of the total capacity and production by 2030.

The Korean government has maintained a consistent national policy for a stable energy supply by fostering nuclear power industries to offset the lack of other energy resources in the country. Nuclear power accounted for

31.3% of the total electricity generation in Korea in 2010 [MEST, 2010]. Since the commencement of the first commercial operation of Kori Unit 1 in April 1978, 21 nuclear power plants (NPPs) are commercially operating as of 2011 with an installed capacity of 18,716 MWe. Four units out of the 21 operating NPPs are pressurized heavy water reactors (PHWRs) at the Wolsung site. The remaining 17 units, located at the Kori, Yonggwang and Ulchin sites, are pressurized light water reactors (PWRs) (Fig. 21.1). There are seven units (three units of OPR 1000, four units of APR 1400) under construction; in addition, six units are in the planning stage of construction.

All nuclear plants are operated by KHNP (Korea Hydro & Nuclear Co.). In addition to the domestic nuclear plant construction, Korea is building four nuclear units of Korean design (APR 1400) in the United Arab Emirates.

In August 2008, the government set out a plan to significantly reduce the nation's dependency on fossil fuels and more than quadruple the use of renewable energy by 2030. In addition, nuclear power will expand to account for 27.8% of total energy consumption in 2030 compared to 14.9% in 2007. The International Atomic Energy Agency (IAEA) officially recognized the Republic of Korea's nuclear transparency by approving the broader conclusion at the regular meeting of the IAEA Board of Governors held in June 2008.



21.1 Current status of nuclear power in Korea, as of June 2012.

21.1.3 The radioactive waste (RAW) management situation in Korea

Spent fuel (SF) generated from nuclear power plants has been stored in spent fuel storage pools at reactors or in on-site dry storage facilities. Dry storage is currently used only for PHWR (CANDU) spent fuel sufficiently decayed for about six years in storage pools. The low- and intermediate-level radioactive waste (LILW) generated from the NPPs has been stored in on-site radioactive waste storage facilities.

Radioactive waste materials are also generated from fuel fabrication processes and they are stored on-site. In addition, the use of radioactive materials in medicine, research work and industry has increased steadily. These facilities are located throughout the country and generate various types of RAW. Radioisotope (RI) contaminated waste from these facilities is stored at an RI waste management facility. There has been much turmoil concerning public acceptance issues associated with the LILW disposal facility site selection, with a number of unsuccessful attempts to select the site.

The Korean government has striven to secure a disposal site for the safe management of RAW since the early 1980s. After a number of failed attempts, the Korean government issued a Public Notice on the selection of a candidate site for the LILW disposal facility, and the city of Gyeongju was selected as the final candidate site in November 2005 following the procedures involving a site suitability assessment, local referenda, etc. as specified in the Public Notice. The Korea Radioactive Waste Management Corporation. (KRMC) was established in 2009 as a new Korean RAW management agency and is currently undertaking the construction of the LILW disposal facility in accordance with the permit issued.

Spent fuel generated from NPPs is stored in the spent fuel storage facility in each unit. The storage capacity for spent fuel has been expanded as a consequence of the delayed construction schedule of the away-from-reactor (AFR) interim storage.

21.2 Radioactive waste (RAW) management strategy, practice and issues

The safe management of RAW is recognized as an essential national task for sustainable generation of nuclear energy and for energy self-reliance in South Korea. Since the early 1980s, the Korean government has attempted to prepare a disposal site for safe management of RAW but failed to secure one due to lack of public consensus and acceptance. In this context, the Atomic Energy Commission (AEC) of the Korean government, the highest decision-making body for nuclear energy policy, approved the 'National

Radioactive Waste Management Policy' at the 249th meeting held on September 30, 1998. This policy stipulated that a LILW facility would be constructed and operated by 2008 and a centralized spent fuel interim storage facility by 2016. The key principles of the national policy on radioactive waste management are as follows:

- direct control by the government
- safety as top priority
- minimization of waste generation
- 'polluter pays' principle
- transparency for site selection process.

However, a revision of the government policy was made at the 253rd AEC meeting on December 17, 2004, after the government failed repeatedly to find a candidate site for the radioactive waste management complex. Therefore, a new government plan for radioactive waste management was announced, basically to separate the sites for the LILW disposal facility and the spent fuel interim storage facility instead of constructing both facilities on one site. The LILW disposal facility is now being constructed in Gyeongju after local referenda. Conversely, the key decision to directly dispose of or recycle spent fuel has not yet been made in Korea. Spent fuel is currently stored at reactor sites under the responsibility of Korea Hydro and Nuclear Power Co. (KHNP), because the 253rd AEC meeting stipulated that the national policy for spent fuel management will be decided later, taking account of domestic and international technological developments.

21.2.1 Sources, types and quantities of radioactive waste

Radioactive wastes arise from the generation of electricity in nuclear power stations and from the use of radioactive materials in industry, medicine, research, and military. There is a wide spectrum of wastes, from those that contain high concentrations of radioactive materials, to general industry and laboratory wastes which are only lightly contaminated with activity.

The Atomic Energy Act (AEA, Article 2.18) of the Republic of Korea defines 'radioactive waste' as radioactive materials or materials contaminated with radioactive materials which are subject to disposal, including spent fuel. The Enforcement Decree of the AEA defines high-level radioactive waste (HLW) as radioactive waste with radioactivity concentration and heat generation over the limiting volume specified by the Ministry of Education, Science, and Technology (MEST). In the strict sense, wastes other than HLW belong to the LILW category in accordance with the AEA. The limiting values on radioactivity and heat generation rate are specified in the MEST Notice No. 2008-31 (Notice of the Standards on Radiation Protection, etc.) [MEST, 2008] as follows:

- radioactivity: ≥4,000 Bq/g for alpha-emitting radionuclides with a half-life of longer than 20 years
- heat generation rate: ≥2 kW/m^3.

The AEA also defines the clearance level adopted from the 'exempt waste' concept of the IAEA radioactive waste classification. The clearance levels in Korea are such that annual individual radiation dose shall be less than 10 μSv/y and the total collective dose below one person-Sv/y concurrently. These are the same as the levels specified in the IAEA Safety Series No. 115 (1996) [IAEA, 1996].

All radioactive wastes are still to be stored in on-site temporary storage until a permanent disposal facility has been constructed. The amount of radioactive waste being stored by April 2012 is 89,865 drums from nuclear power plants (KHNP, 2012). (Hereafter, 'drum' means '200-liter drum equivalent' unless otherwise stated.) The total capacity of temporary storage in NPP sites is 109,900 drums and the accumulated radioactive waste stored at each NPP site is around 77.7% of their storage capacity, as shown in Table 21.1. Although the volume of waste arising from radioisotope use is still relatively small compared to power reactor waste volume, the annual generation rate is expected to rise rapidly as industrial use of radioisotopes increases. The waste type and volume of LILW is shown in Fig. 21.2.

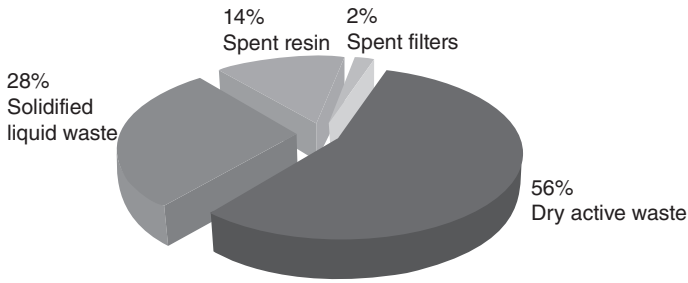
21.2.2 Radioactive waste treatment

To ensure its safe discharge into the environment, liquid radioactive waste has to fulfill very strict requirements connected with the limits of radioactive substances and other impurities (suspended particulates, chemical, biological, heavy metals, etc.). To achieve the standards described in national

Table 21.1 The status of the LILW storage in nuclear power stations (as of April 2012)

Nuclear power stations		Storage capacity (no. of drums)	Cumulative amount (no. of drums)
Location	Number of reactors		
Kori	4	50,200	41,012
Yonggwang	6	23,300	21,601
Ulchin	6	18,929	16,020
Wolsong	4	13,240	10,987
Shin-kori	1	10,000	245
Total		115,669	89,865

Source: KHNP (2012) from http://www.khnp.co.kr.



21.2 The composition of LILW waste generated in Korea, as of 2005 (Ahn *et al.*, 2009).

regulations, radioactive waste has to be treated, including volume reduction and reduction of radioactive compounds and other solutes in the effluent.

NPPs currently in operation in Korea have their own gaseous, liquid, and solid waste treatment facility and on-site storage facilities to ensure the safe management of RAW generated in the process of operation. The gaseous waste treatment system comprises gas decay tanks and/or charcoal delay beds. The liquid waste treatment system is equipped with either liquid waste evaporators or selective ion exchangers. The solid waste treatment facility has spent resin drying systems, spent filter processing and packaging systems, concentrated waste drying systems, and dry waste compactors. The RI waste generated from domestic medical research, industrial RI users, and research institutes is collected and stored at the Central Research Institute (CRI) of KHNP in Daejeon. Around 90% of LILW comes from NPP and the rest arises from industry, medicine, and research institutes.

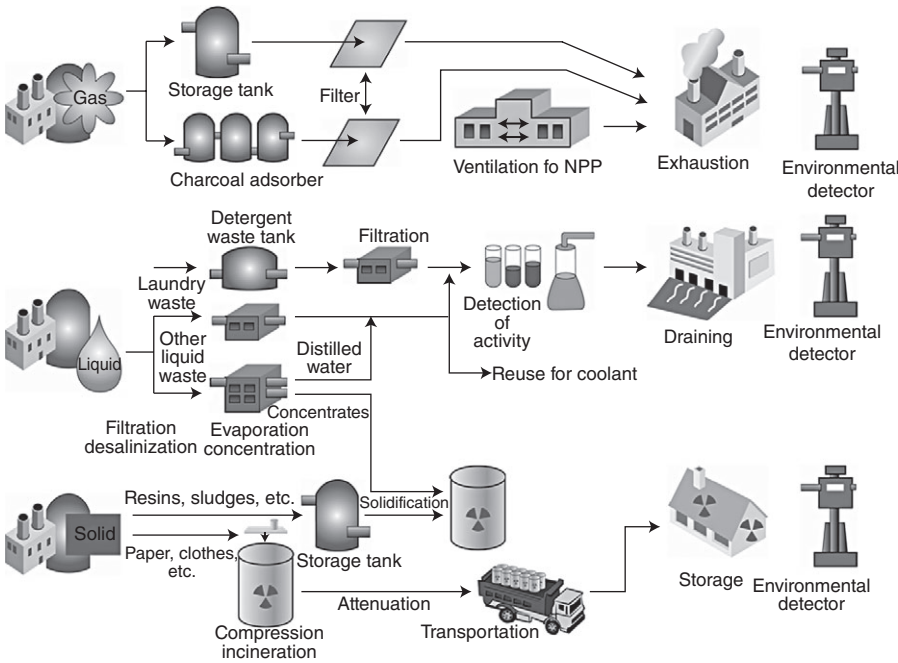
Generally, the type of LILW is classified as follows:

- power plants: dry active waste, spent resin, spent filter, and concentrated waste
- non-power plant sources (RI waste): dry active waste (combustible or non-combustible), hepatitis waste, organic liquid waste, spent sealed source, spent resin, spent filters, and concentrated waste.

Figure 21.3 summarizes the process steps for treatment of solid, liquid and gaseous wastes in Korea.

Solid radioactive waste (SRAW)

Most SRAW consists of dry active waste (DAW) and secondary process waste. The DAW is generated during maintenance and repair of contaminated systems and includes items such as used parts, paper, clothes, gloves and shoes. Secondary waste is generated from the liquid RAW treatment system and includes concentrated wastes from evaporators, spent resin from demineralizers, and spent filters from liquid purification systems.



21.3 Process steps for radioactive waste treatment.

The DAW is compressed by a conventional compactor (capacity: 2,000 tons) into 200 L drums. Solidification by Portland cement, which had been commonly applied in the past, is no longer used. Instead, the concentrated waste is now dried and stabilized by paraffin wax in drums, and spent resin is kept in a high-integrity or equivalent container after drying in the spent resin drying facility. Spent filters are stored in shielded high integrity containers (HIC).

Liquid radioactive waste

Liquid RAW can be divided into process drains, floor drains and laundry drains based on the sources of waste generation. It is mainly generated from the clean-up and maintenance processes of reactor coolant and related systems containing radioactivity. In general, liquid RAW is treated with evaporators, demineralizers, and/or filters. The effluent is released to the sea after monitoring whether the radioactivity of liquid effluent is lower than regulatory limits. It is also common for liquid wastes to be treated with ultracentrifugation, ion exchange, and reverse osmosis.

The Ministry of Education, Science, and Technology (MEST Notice No. 2008-31) prescribes the effluent control limit (ECL) for liquid effluent being discharged into the environment at the restricted area boundary. Operators

must conduct periodic assessments for the expected off-site dose due to the liquid effluent discharged into the environment, and routinely report results to the regulatory body (Korea Institute of Nuclear Safety, KINS).

Gaseous radioactive waste

Gaseous RAW is mainly generated from degassing of the primary system and ventilation systems in the radiation controlled area of NPPs. Gaseous waste from the primary system is treated by gas decay tank or charcoal decay bed to reduce radioactivity, and released into the atmosphere through a radiation monitor. Gaseous waste from the building ventilation system is also exhausted under continuous monitoring through high-efficiency particulate (HEPA) and charcoal filters into the environment.

The MEST addresses the maximum radioactivity concentration, ECL, for gaseous effluent being released into the atmosphere at the restricted area boundary (MEST Notice No. 2008-31). The licensee must conduct a periodic evaluation of the anticipated off-site dose due to gaseous effluent released into the environment, and routinely report results to the KINS. The Enforcement Decree of the AEA and the MEST Notice No. 2008-31 (Standards on Radiation Protection, etc.) prescribe discharge limits of gaseous and liquid radioactive effluents to be released from nuclear facilities into the environment, along with annual dose constraints of the population living around nuclear facilities.

In practice, nuclear facilities are operated with targets which are more restrictive than the discharge limits. In addition, some facilities also apply the derived release limits based on a small fraction of the dose limits for convenience for a field application. Whether related limits are met is verified by periodic inspection or the examination of regular reports submitted to the regulatory body.

The radiation dose and its effect on individuals around nuclear facilities are assessed monthly by using the *Off-site Dose Calculation Manual* (ODCM, Reg. Guide 1.109) [US-NRC, 1977]. The assessments are based on the radioactivity of released liquid and gaseous effluents, atmospheric conditions, metabolism, and social data including agricultural and marine products of the local community within a radius of 80 km.

The Korea Atomic Energy Research Institute (KAERI) in Daejeon and KHNP carry out R&D on RAW management. Treatment and disposal of HLW/SF is studied by KAERI. KHNP studies the treatment and disposal of LILW and interim storage of spent fuel. Technological developments are currently focused on the following topics:

- waste treatment and volume reduction technology
- low-level waste vitrification technology

- LILW disposal and safety assessment technology
- improvement of existing technology for spent fuel storage and transportation, and development of advanced spent fuel storage technology.

In addition to current use of conventional treatment methods such as evaporation, compaction, drying and cementation, advanced technology for LILW treatment is being developed. Vitrification has been identified as the most promising innovative technology from the point of view of being environmentally sound and of being able to substantially reduce the volume of LILW, to improve the waste stability and to enhance the public acceptance of its disposal. Vitrification immobilizes the radionuclides in a stable solid glass form and the associated volume reduction should result in efficient and prolonged use of a repository, which is most important for a small, densely populated country.

A feasibility study of the vitrification process was initiated in 1994 and a pilot-scale vitrification facility was installed in July 1999. This facility consists of an induction heater, cold crucible melter (CCM) for combustible waste, a plasma torch melter (PTM) for non-combustible waste, and an off-gas treatment system. KHNP's research center (CRI) located in Daejeon has developed the technology with a target for commercialization of the process from 2005. The Ulchin Vitrification Facility (UVF) is the world's first commercial facility for the vitrification of LILW generated from NPPs using CCM technology. The construction of the facility began in 2005 and was completed in 2007. From December 2007 to September 2009, all key performance tests, such as the system functional test, the cold test, the hot test, and actual waste testing, were performed successfully. The UVF started commercial operation in October 2009 for the vitrification of LILW waste (Jo *et al*., 2010).

21.2.3 Radioactive waste disposal

Since the creation of the legal grounds for the implementation of the project by the 1986 revision of the Atomic Energy Act (AEA), the Korean government has actively implemented the selection of the sites for radioactive waste disposal facilities. There have been nine failed attempts to secure a disposal site from 1986 to 2004 due to:

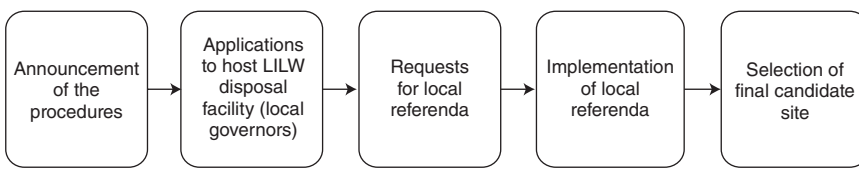
- safety concerns about the disposal facility,
- lack of transparency and fairness during project implementation,
- lack of social consensus among the stakeholders.

In February 2004, the Ministry of Knowledge Economy (MKE) announced new site selection procedures, and MKE/KHNP made various efforts to enhance the acceptance by local residents of disposal facilities. As a result,

local residents voluntarily petitioned to host the facilities in ten areas, but site selection ultimately halted due to the absence of preliminary applications by local government leaders. In March 2005, MKE organized the Site Selection Committee (SSC) in order to guarantee the transparency and fairness of the site selection process. The SSC, consisting of 17 civilian experts from diverse fields, managed and supervised the entire site selection process. In addition, the 'Special Act on Support for Areas Hosting Low and Intermediate Level Radioactive Waste Disposal Facilities' (MKE Notice No. 2005-146) was legislated and announced in March 2005 to stipulate support for areas hosting LILW disposal facilities, including special financial support, entry fees, and relocation of the KHNP headquarters. The act also stipulated the following to enhance the democracy and transparency of the selection process:

- the host area was to be selected through resident voting in accordance with the Referendum Act,
- the selection plan, site survey results, and selection process were to be implemented openly and transparently,
- open fora and discussions were to be held for local residents.

Accordingly, in June 2005, the MKE announced the candidate site selection method and procedures as well as the support to be provided to the host areas and initiated the process through an announcement regarding LILW disposal facility candidate site selection. Regarding candidate site selection procedures, as shown in Fig. 21.4, the local governors must apply to host the facilities with consent from local councils. Then, in accordance with the results of the site suitability assessment, the MKE requested local governors to conduct local referenda in appropriate regions as required by the Referendum Act. Local governors proposed and held the referenda. Based on the results of local referenda, areas with the highest percentage of favorable responses were selected as the final candidate sites. Local governments that had appropriately applied to host the LILW disposal facility by August 31, 2005 were in the four areas of Gunsan, Gyeongju, Pohang, and Yeongdeok County, and these four local governments conducted referenda. In accordance with the results of local referenda (Table 21.2), the city of Gyeongju was selected as the final candidate site (MKE Notice No. 2005-133).



21.4 Site selection procedures of the LILW disposal facility.

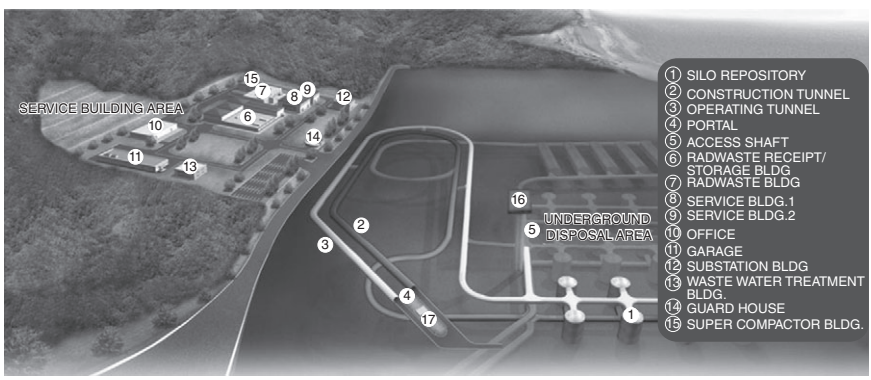
The area of the disposal site accommodates a total of 800,000 drums of LILW, and, as the first stage of construction, a rock cavern type of repository for up to 100,000 drums was chosen. However, the disposal method for further expansion will be decided depending on the nature of the site condition. The disposal facility to be constructed in Gyeongju was named 'Wolsong Low- and Intermediate-level Radioactive Waste Disposal Center' operated by KRMC under the jurisdiction of MKE which was established on January 1, 2009 (Figs 21.5 and 21.6). As of June 2012, the disposal facility is almost 90% complete (Fig. 21.7) and the date for initial operation is mid-2014, taking into account the construction period.

In the main review phase, after completion of three rounds of Q&A, a few key technical issues (KTIs) were brought out and profiled for further intense deliberation. The KTIs that needed to be taken into consideration throughout the later part of the main review phase can be summarized as follows:

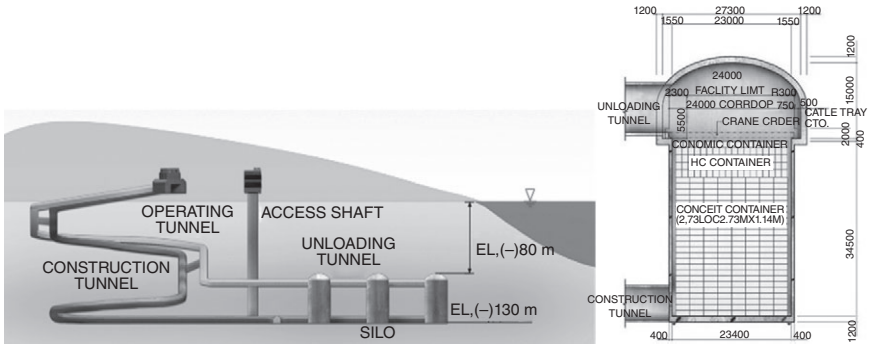
Table 21.2 Results of referenda for site selection in 2005

Classification	Gyeongju	Gunsan	Yeongdeok	Pohang
No. of eligible voters	208,607	196,980	37,536	374,697
No. of voters (absentees)	147,636 (70,521)	138,192 (65,336)	30,107 (9,523)	178,586 (63,851)
Voter turnout	70.8%	70.2%	80.2%	47.7%
Vote for	89.5%	84.4%	79.3%	67.5%

Source: Park *et al.*, 2009.



21.5 View of the Wolsong LILW disposal center.



21.6 Cross-section view of the underground facilities in the LILW repository.

21.7 Construction of the LILW repository (87% complete, as of June 2012; KRMC, 2012).

- groundwater infiltration rate into silos: re-estimation of the groundwater infiltration rate into the concrete
- silos during the post-closure phase, in combination with justification of the human intrusion scenarios
- quality control of geochemical data: reconfirmation of the representativeness of empirically determined site-specific geochemical data (e.g. sorption coefficients, diffusion coefficients, etc.)
- long-term management of uncertainties in geochemical data
- seismic safety and design: verification of the geological structure model and tectonic activity of the site
- structural stability of the rock caverns and silos.

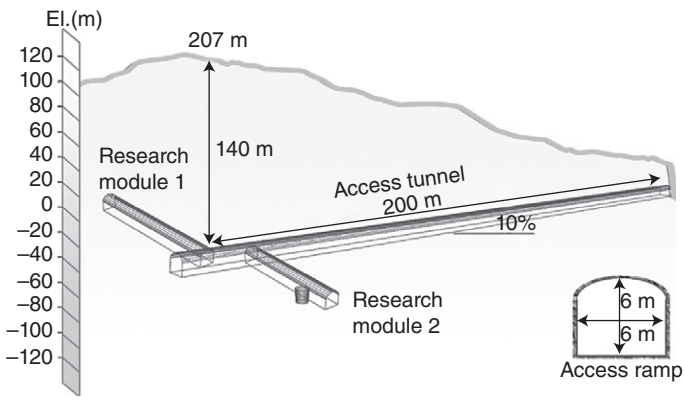
The above KTIs were resolved through regulatory dialogues and requests for more detailed information along with the applicant's amendments to the license application documents, reflecting the results of further supplementary site surveys, safety assessments, and design changes, which occurred during the review process.

KAERI underground research tunnel (KURT)

A small-scale underground research laboratory, KAERI Underground Research Tunnel (KURT) at KAERI in Daejeon, was constructed to develop a Korean disposal system for the HLW repository, including spent fuels, between March 2005 and November 2006. The KURT, with an access tunnel and two research modules, as shown in Fig. 21.8, is located in a mountainous area inside the KAERI territory. The KURT, has a total length of 255 m with a 180 m long access tunnel and two research tunnels 75 m long in total. The maximum depth of 90 m could be effectively achieved by selecting the tunnel direction to the peak of a mountain. The horseshoe shaped tunnel, 6 m wide and 6 m high, is located in a granite rock body (Fig. 21.8). Regardless of limited applications of KURT, which only handles naturally occurring radionuclides, the KURT facility will be a major infrastructure for validating the safety and feasibility of the suggested disposal system by various *in-situ* experiments:

1. Single hole heater test in rock.
2. THM (thermal-hydraulic-mechanical) behavior of engineered barrier systems (EBS).
3. EDZ (excavation disturbed zone) characteristics and mechanical stability of rock.
4. Retardation of solute migration through fractured rock.
5. Site investigation techniques.
6. Hydrogeological and geochemical baseline data (Kwon *et al.*, 2009).

The current 10-year plan for mid- and long-term nuclear R&D on HLW disposal was accepted by the AEC in 1997. This plan includes a program for development of a Korean repository for HLW disposal and for the associated system performance assessment. After completion of the



21.8 Schematic internal configuration of KURT [Cho *et al.*, 2007].

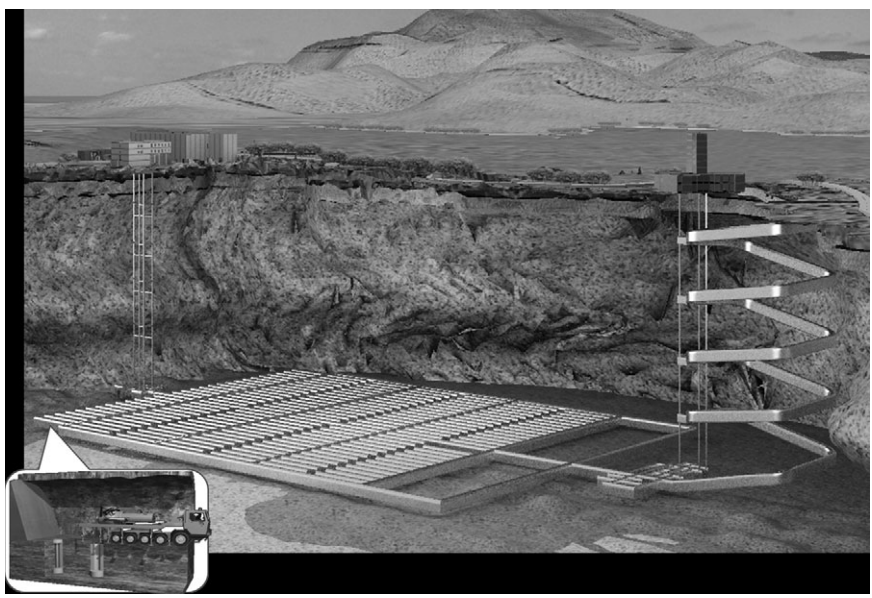
combined research output of this 10-year study, the Korean government will define the direction and prioritization of further R&D activities for HLW disposal. Since 1997, KAERI has been developing a permanent disposal facility for HLW and a total system performance assessment (TSPA). Its current R&D activities are focused on the preliminary conceptual design of the Korean Reference Disposal System (KRS), development of the key technologies, and geo-environmental studies to confirm the KRS's safety, as shown in Fig. 21.9. Currently, the four major projects underway at KAERI are:

1. repository system development;
2. a TSPA;
3. geo-environmental science research; and
4. construction and operation of a KAERI underground research tunnel (KURT) to demonstrate the KRS's performance relevant to the functional criteria established in the disposal concept (Fig. 21.8).

21.3 Spent fuel management strategy, practice and issues

21.3.1 Spent fuel inventory

Spent fuels can be categorized into those from commercial NPPs and those from research reactors. Spent nuclear fuels from commercial NPPs are



21.9 Korean Reference Disposal System.

stored on site in spent fuel storage (water) pools or in a dry storage facility. All the spent fuels from the 17 PWRs in Korea are stored in pools on site. About half of the spent fuels from the four CANDU reactors is stored in pools and the other half is stored in dry silos or dry casks on site. As of September 2011, 5,408 tons of spent fuel from PWRs and 6,431 tons of spent fuel from CANDU reactors are stored at four sites: three sites for PWRs, one site for CANDUs (NSSC, 2011). The annual addition to the amount of spent fuel is about 690 mtu. After 2045, spent fuel stores from the CANDU reactors will be full because of decommissioning of the CANDU reactors. The capacities, inventories and types of spent fuel in storage are given in Table 21.3 (NSSC, 2011).

Spent fuel and irradiated fuel from the HANARO research reactor are stored in the storage pool on site at the Korea Atomic Energy Research Institute (KAERI). Up to 20 PWR fuel assemblies can be stored in the storage pool after irradiation tests. As of September 2011, 4 tons of spent fuel from HANARO was stored in the pool on site (NSSC, 2011). HANARO is a multi-purpose research reactor used for fuel performance testing, material irradiation testing, radio isotope (RI) production, and basic science and applications studies.

21.3.2 Spent fuel storage

Spent fuel generated from NPPs is stored in the spent fuel storage facility in each unit. The storage capacity for spent fuel has been expanded as a consequence of the delayed construction schedule of the away-from-reactor (AFR) interim storage in accordance with the conclusions of the 249th and the 253rd meetings of the AEC. Taking into consideration the sufficiency of spent fuel storage capacity beyond 2016, the national policy for spent fuel management, including the construction of the interim storage facility for spent fuel, shall be decided in a timely manner through national consensus by public consultation among the stakeholders.

To expand the spent fuel storage capacity, the utility company, Korea Hydro & Nuclear Power Co. (KHNP), is installing high density storage

Table 21.3 Spent fuel storage (as of September 2011)

NPP Site	Type of storage	Capacity* (mtu)	Inventory (mtu)
Kori	Wet	2,472	1,869
Yonggwang	Wet	2,686	1,949
Ulchin	Wet	2,328	1,591
Wolsong	Wet and dry	9,441	6,431
Total		16,927	11,839

* Except emergency core.

racks, transferring spent fuel between units and building dry storage. High density storage racks have been installed in Kori 3 and 4, Ulchin 1, 2, 3 and 4, and Yonggwang 3 and 4. Dry storage facilities have been installed on the Wolsong site for Wolsong 1, 2, 3 and 4 units which are CANDU reactors. By adding 100 canisters in 2006, 300 canisters are installed on site. In addition to the canisters, seven modules of MACSTOR (Modular Air-Cooled STORage)-400 with 3,175 mtu total capacity have been installed and in operation since May 2010.

The spent fuel storage pool of the HANARO reactor is a heavy concrete structure, lined with stainless steel plate. The vault comprises three storage lattices. The vault has enough capacity for temporarily storing new fuel as well as spent fuel to be generated during normal operation of HANARO for 20 years.

The Korean government has striven to secure a spent fuel management site since the early 1980s. However, the national policy for spent fuel management including construction of the centralized spent fuel interim-storage facility was to be decided in view of domestic and international technology developments later on. The national policy for spent fuel management will be decided later in consideration of domestic and international technology developments. Reprocessing activities have not been conducted in Korea.

21.3.3 Advanced fuel cycle to address spent fuel issues

The international nuclear community recognizes the potential of nuclear energy systems to cope with increasing energy demand and international protocol for climate change even after the Fukushima accident. International cooperative programmes have been initiated to develop new systems that secure stable energy supply and have improved public acceptance, safety, and cost-effectiveness. The Republic of Korea is actively participating in these programmes currently, such as the Generation IV International Forum (GIF) and the International Project on Innovative Nuclear Reactors and Fuel Cycle (INPRO).

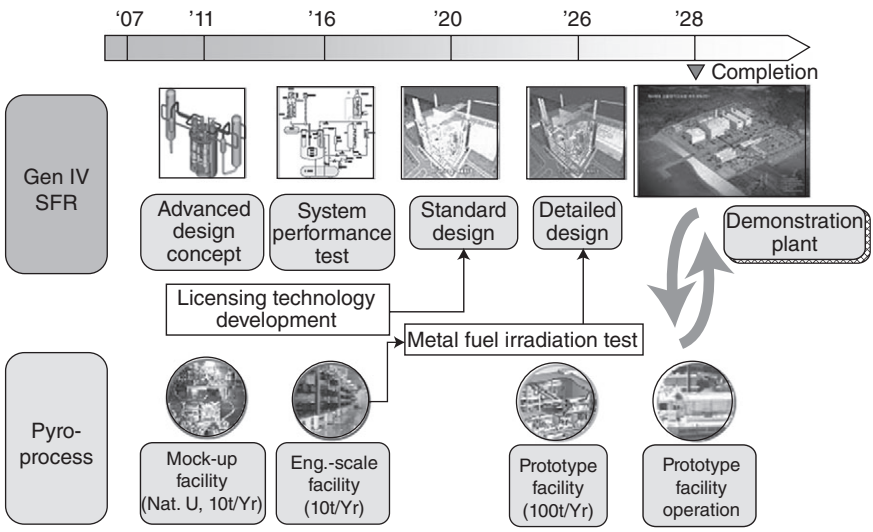
Korea has been a chartered member of GIF since 2000 and plays a significant role in the development of Gen-IV. GIF was organized for collaborative development of new generation nuclear energy systems aiming for 2030 that can be accepted by the public and the energy market with excellent technical features and competitive economics, with 13 members leading nuclear utilization and development in the world taking part in GIF. GIF selected six systems of the most promising concepts as the Generation IV nuclear energy systems (Gen-IV) in 2002 and has been conducting collaborative R&D for each system through multilateral agreements since 2005. Korea focuses on SFR (sodium-cooled fast reactor-see Fig. 21.10) and

VHTR (very high temperature reactor) among the six Gen-IV systems. SFR is expected to use and recycle uranium resources effectively and minimize high-level radioactive waste with proliferation resistant fuel cycles. Korea is participating in six collaborative projects, tackling safety and operation, advanced fuels, and component design and balance of plant in SFR. Korea's Long-term Development Plan for Future Nuclear Energy Systems, approved in December 2008, also presents a milestone and deliverables of SFR and pyro-processing technology.

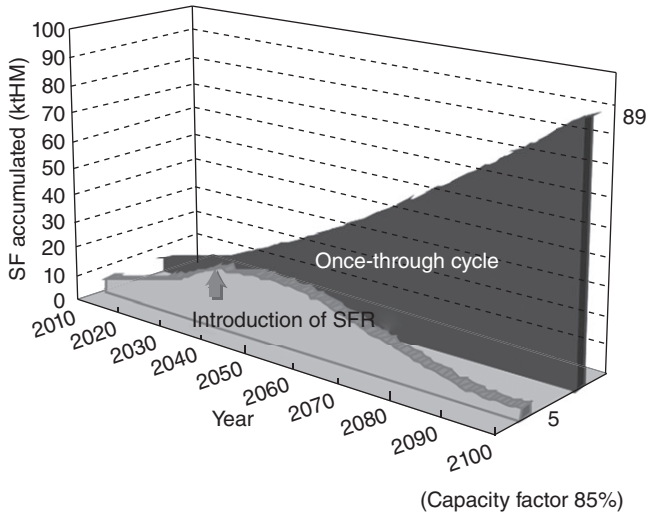
KAERI has been developing pyro-processing technology (Fig. 21.10) for recycling useful resources from spent fuel since 1997. The process includes pre-treatment, electro-reduction, electro-refining, electro-winning, and a waste salt treatment system. The removal of transuranic elements (TRU), Cs, and Sr from spent fuel allows the repository burden to be reduced by a factor of 100, compared with the case without removal. Fission products (FP) are recovered and transferred to a repository. As a result of pyro-processing, both repository efficiency and uranium usage are increased up to 100-fold with strong proliferation resistance.

According to the analysis of KAERI, spent nuclear fuel stock at the end of this century can be maintained at a level lower than that of today by introducing SFRs coupled with pyro-processing technology in the 2030s (Fig. 21.11).

Korea has had an open fuel cycle, without reprocessing in compliance with the terms of its nuclear cooperation agreement with the USA, which



21.10 The Republic of Korea's long-term development plan for future nuclear energy systems (Kim, 2010).



21.11 Cumulative PWR spent fuel (Kim, 2010).

needs to be renewed in 2014. In 2008, the IAEA approved an electro-refining laboratory – the Advanced Spent Fuel Conditioning Process Facility (ACPF) at KAERI which is to be built by 2011 and expanded to engineering scale by 2012. This is envisaged as the first stage of a Korea Advanced Pyro-processing Facility (KAPF) to start experimentally in 2021 and become a commercial-scale demonstration plant in 2025. In connection with renewal of the US-ROK agreement in or by 2014, discussions are proceeding on pyro-processing.

21.4 Decommissioning and decontamination (D&D) strategy, practice and issues

Korean decommissioning and decontamination (D&D) work on the retired research reactors KRR-1 and 2 and the uranium conversion facility (UCF) at KAERI is under way. Hundreds of tons of metallic and concrete wastes are expected from the D&D of these facilities. Therefore, countermeasures are being taken to deal with the amount of waste generated by dismantling these retired nuclear facilities. Recycling or volume reduction of the large quantities of metallic and concrete wastes are key waste management options due to the difficulty in securing a waste disposal site in Korea and the capacity limitation of the temporary waste storage facility at KAERI. Recycling or volume reduction through application of appropriate treatment technologies has merits from the viewpoint of resource recycling as well as a decrease in the amount of waste to be disposed of resulting in reduced disposal cost and enhanced disposal safety.

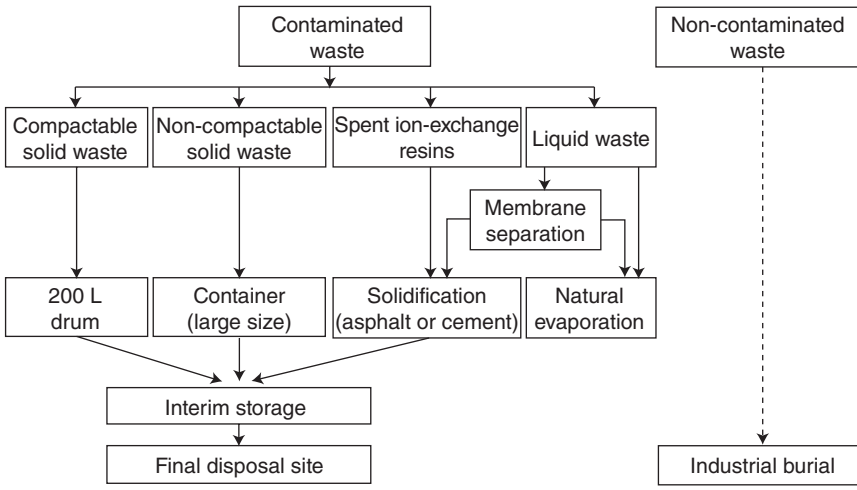
21.4.1 D&D of TRIGA Mark-II and III research reactors

TRIGA Mark-II, the first Korean research reactor (KRR-1), started operation in 1962, and the second, TRIGA Mark-III (KRR-2) located in Seoul, has been operational since 1972. These two research reactors, located at the former KAERI site in Seoul, were permanently shut down at the end of 1995. As a replacement for the TRIGA research reactors, the 30 MW_{th} multipurpose HANARO research reactor was constructed in 1995 located at KAERI in Daejeon and has operated successfully since then. The D&D of KRR-1 and 2 research reactors was started in January 1997. The decommissioning plan, environmental impact assessment and decommissioning design were carried out in 1998. In July 1998, all SF from the TRIGA Mark-II and III reactors was safely transported to the US. At the end of 1998, the decommissioning plan was submitted to the Ministry of Education, Science, and Technology for licensing, and the Korea Institute of Nuclear Safety (KINS) reviewed it in 1999. The report of their review was considered in January 2000 by the Expert Group for Environmental Radiation, one of the four groups of the Nuclear Safety Commission, and the recommendation made by that Expert Group was submitted to the Commission for its final approval. At the moment, KRR-2 has been completely dismantled, whereas the decommissioning of KRR-1 was started in 2011 and will be completed by the end of 2014.

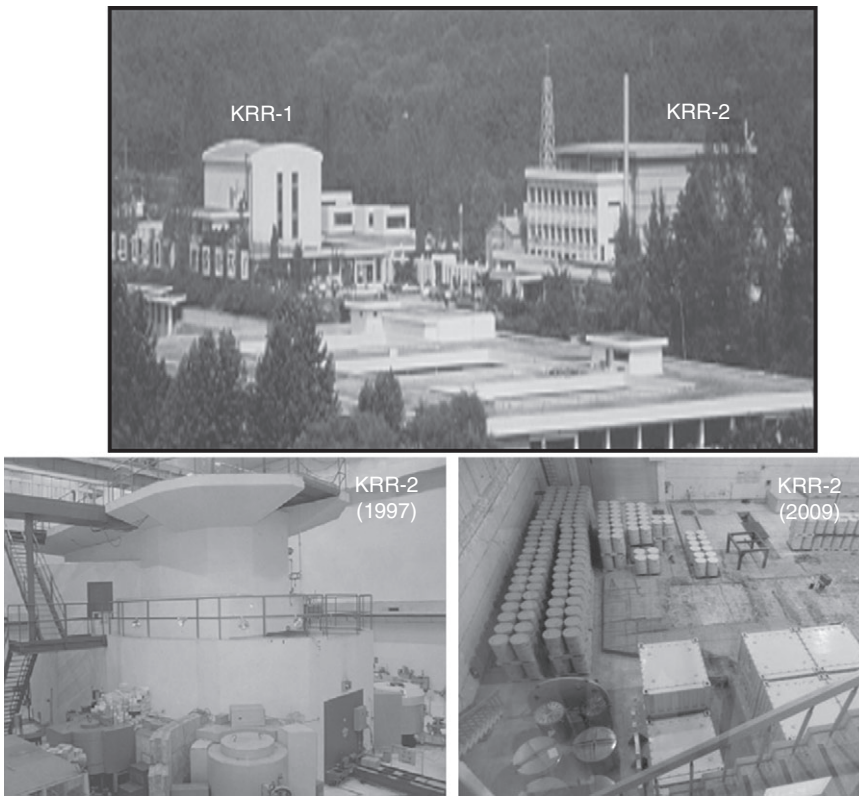
Radioactive wastes from the decommissioning of KRR-1 and 2 were classified according to their characteristics and radioactivity levels, packed into 200 L drums or 4 m^3 containers and stored in the reactor hall of the KRR-2 according to the process scheme of radioactive waste treatment from decommissioning sites, shown in Fig. 21.12 Radioactive waste generated from KRR-1 and 2 contains ^{60}Co and ^{152}Eu as major radionuclides in the activated waste and ^{60}Co and ^{137}Cs in the case of the contaminated waste. The current status of KRR-1 and 2 is shown in Fig. 21.13, and complete D&D of both will be performed within a few years later.

21.4.2 D&D of uranium conversion facility (UCF)

The uranium conversion facility (UCF) located at KAERI was operated from 1982 to 1992. After the localization of nuclear fuel fabrication technology, it was shut down in 1993. UCF decommissioning began in 2001 and radioactive waste from UCF has been stored in a temporary storage building in the conversion facility. All the wastes are contaminated mainly with natural uranium. Currently, the dismantling of 26 out of 27 rooms at UCF has been conducted (Fig. 21.14), including decontamination of concrete surfaces, removal of contaminated soil, and completion of treatment of sludge waste in a lagoon.



21.12 Procedures for the treatment of contaminated wastes.



21.13 KRR-1 and 2 and decommissioning status of KRR-2.

21.14 Decommissioning of UCF.

Research achievements to date are:

- development of volume reduction technology for large amounts of radioactive concrete wastes
- development of soil decontamination technology for remediation of nuclear sites after decommissioning
- development of melting technology for decontamination of a hundred tons of slightly contaminated metallic wastes generated from KRR-1 and 2 and UCF
- development of technologies for safe management of irradiated graphite arising from decommissioning of KRR-1 and 2
- development of a database system for management and data assessment from D&D activities
- development of chemical decontamination technology applicable to metal wastes contaminated with UN (uranium nitride), AUC (ammonium uranyl carbonate), and UO_2 generated by dismantling UCF
- development of the safety assessment methodology of the decommissioning process
- simultaneous remote measurement of alpha/beta contamination in highly contaminated facility

- decontamination technology development
- waste treatment technology development.

Major R&D activities are now concentrated on development of the decommissioning waste reduction and recycling technology for commercial NPPs and nuclear facilities.

21.5 Conclusion

Given the scarcity of Korea's primary energy resources, nuclear power is vitally important as an engine of growth for the nation. Korea has followed a set of consistent policies and executed steady plans to expand nuclear power. With a significant share of nuclear power in the energy mix, the disposal of RAW and SF is looming large as a high-visibility national issue. A low-and intermediate-level waste disposal site has been selected and the facilities are currently under construction with its full operation expected in 2014. Spent fuel management has also become imminent. Although no satisfactory resolution is in sight in the foreseeable future, various options are being studied with the government's keen interest and full support. Korea has also designed a rigorous process for decontaminating waste materials.

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22

China: experience of radioactive waste (RAW) management

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Abstract: Progress in the management of China's radioactive waste (RAW) is described, including waste generation, waste management policy, and current practices in regional disposal of low and intermediate level waste (LILW) and development of a geological disposal facility for hight level waste (HLW).

Key words: China radioactive waste management, geological repository, regulations and policies.

22.1 Introduction

China started its commercial nuclear industry in the early 1970s; however, development was slow prior to 2000. To meet the energy demands of its rapid economic growth and social development over the last 30 years, China has been building an electricity supply system with multiple sources. Coal-powered electrical plants still play a major role. Meanwhile, cleaner energy, including nuclear, will see significant growth considering factors of resource, transportation, environmental concern and climate change.

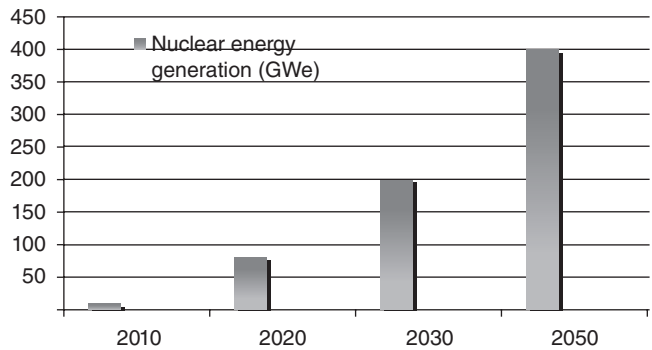
22.1.1 Background

Up until 2011, China's nuclear power was still very small compared with other major world powers and only ~1.5% of the nation's electricity was generated by nuclear power. China has 12 operating nuclear power units (Table 22.1), distributed along coastal areas. Plate V (between pages 448 and 449) shows the geographical distribution of nuclear power plants (NPPs) in China.

With current worldwide interest in nuclear power as a clean energy source and the technical development of waste management and disposal in China, nuclear is becoming a significant proportion of China's power generation. As of June 2010, the official nuclear capacity targets were 80 GWe by 2020, 200 GWe by 2030 and 400 GWe by 2050 (Fig. 22.1). The aim

Table 22.1 China's operating nuclear power reactors

Plant/reactors	Location	Size (per reactor)	Reactor	Company	Operation start date
Daya-Bay (2)	Shenzhen (GD)	940 MWe	PWR	CGNPC	1994
Qinshan I (1)	Hangzhou-bay (ZJ)	280 MWe	PWR	CNNC	1994
Qinshan II (2)	Hangzhou-bay (ZJ)	610 MWe	PWR	CNNC	2002–2004
Qinshan III (2)	Hangzhou (ZJ)	670 MWe	PHWR	CNNC	2002–2003
Ling-Ao (2)	Guangdong (GD)	935 MWe	PWR	CGNPC	2002–2003
Tian-wan (2)	Jiangshu	1,000 MWe	PWR (VVER)	CNNC	2007
Ling-ao II (1)	Guangdong (GD)	1,040 MWe	PWR	CGNPC	2010–2011
Total reactors	12				



22.1 China's nuclear power goals up to 2050.

is that by 2050, the nuclear electricity generated should reach around 15–25% of overall electricity generated in China, similar to other superpowers [1–4].

China also has 12 research reactors, 2 uranium enrichment facilities in Gansu, 3 major research facilities mainly in Beijing, and also 32 storage facilities and 2 low and intermediate level waste disposal facilities (LILW) for dealing with the waste from past military and general research reactors, as well as for covering the waste from the newly built coastal NPP. The inventory from one of the waste facilities (in Gansu Province) is given in Table 22.2.

Table 22.2 Waste inventory from Gansu radioactive waste facility

No.	Disposal options	Origin	Volume (m^3)	Percentage of total radioactivity
1	Long-term storage	Seriously contaminated equipment and spent radioactive sources	3	70.9
2	Milling pond	Sludge	15	7.5
3	Mining backfilling	Contaminated equipment and soils	8,747	18.3
4	Tailing dam	Contaminated soil	5,200	2.7
5	Drainage backfilling	Building rubbish	2,450	0.1
6	Residual			0.6
Total			16,415	100

The recent surge in nuclear power has brought much attention to China's overall nuclear programme and the concerns are mainly in the following areas:

- social and economic impacts of nuclear energy,
- the large capital investment required,
- reactor central control systems, including plant safety, radiation protection and emergency accidents, lack of qualified trained engineers and workers, lack of advanced technology,
- uranium mine resources plus management, and, in particular,
- waste management and repository resources.

22.1.2 Recent developments

China's nuclear programme is very ambitious. In 2009, China began to construct six NPP, and construction of 11 more began in 2010. It is planned to build 52 new reactors over the next five years, although after Fukushima a hold was placed on new licence applications and the programme has slowed. Meanwhile, the volume of China's radioactive waste (RAW) is predicted to increase up to 10-fold by 2020, mostly from its 80 GW capacity new build plan. A £10 billion investment in research into radioactive waste management and repository investigation has been included in the next National 5–10 Year Plan in Science & Technology for the overall national energy programme in China.

China is also developing its own fast neutron reactor, with some significant breakthroughs in Generation IV reactors. In particular, China's Experimental Fast Reactor (CEFR) developed by the China Institute of Atomic Energy and Nuclear Power Research Institute under China National

Nuclear Co. (CNNC) achieved criticality in July 2010, making China the eighth country to develop fast reactor technology. This fast reactor project uses sodium as a coolant to generate 65 MWe (thermal) and 20 MW (electric) power and has been financially supported by China's National 863 Research Programme.

22.1.3 Radioactive waste management and regulations in China

The Chinese government and research communities have also paid attention to the issues of radioactive waste disposal and repository siting and design. Waste generated from NPP is, for the most part, currently stored at the NPP sites where the wastes are generated as well as at research institutions that have reactors. The accumulated low and intermediate level radioactive waste (LILW) will eventually be sent to near-surface disposal facilities. The high level wastes (HLW) will be sent to a geological repository when it is available; this is expected to be sometime around 2050 [5–7].

Most of China's regulations and standards are developed based on international safety standards in combination with the Chinese situation. China's current spent fuel (SF) management policy is to reprocess. However, the SF generated so far is still in interim storage, either at or away from the reactors. China's radioactive waste policy serves as a baseline for China's radioactive waste regulations, which are in place to guarantee that there will be no radioactive waste burden left for future generations.

Adapted from the IAEA regulations [8], waste producers in China must:

1. Minimize waste in fuel production and fuel cycles, materials classification and purification.
2. Guarantee a high volume reduction.
3. Use high quality waste packaging materials along with safety regulations that cover transportation and *in-situ* storage during periods when the waste may be exposed.
4. Centralize and control disposal and control release from a waste package which includes enhanced monitoring.
5. Design, construct and operate all facilities and practices for radioactive waste within these practices.

The liquid RAW generated at NPP must be immobilized and solidified. The regulations mandate that the implementers of waste disposal must be relatively independent from the waste producers. The waste disposal service is not chartered by or sponsored by the central government. Five regional sites in Guangdong Province, Zhejiang Province, Gansu and Beijing have been built, mainly for disposal of LILW. For HLW, including SF, current

practice is to temporarily store the wastes, while the liquid RAW is being solidified.

In terms of the legislative framework, the China Atomic Energy Authority/Agency (CAEA) is responsible for development of policies concerning the peaceful uses of nuclear energy:

- development of industry standards;
- control of nuclear materials;
- acting as a leading body for nuclear accident response, in particular for organizing the State Committee of Nuclear Accident Coordination;
- reviewing and approving the nuclear energy development project;
- reviewing and approving R&D projects.

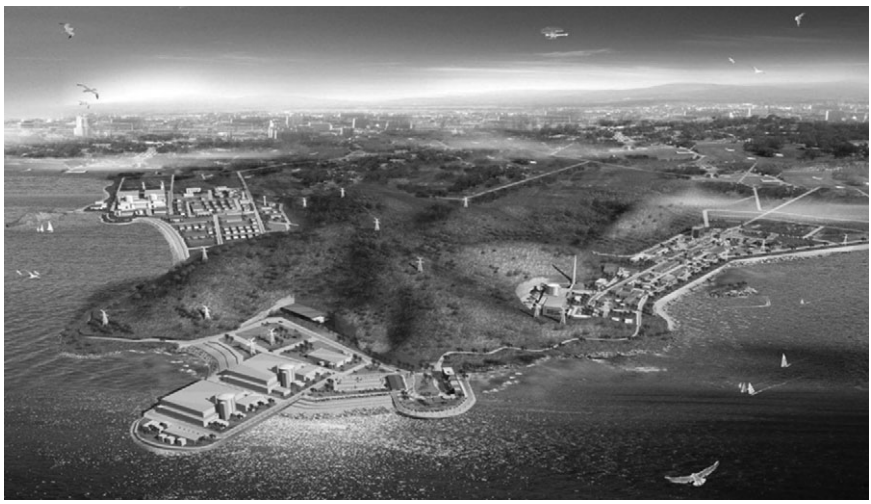
The People's Congress developed the Regulatory Framework Act to address some issues with the signed Presidential Regulations. The State Council is responsible for the promulgation of regulations, which are issued with the signature of the Prime Minister.

22.2 Sources, types and classification of waste

As specified in the Law of the People's Republic of China on Prevention and Control of Radioactive Pollution [9], RAW is defined as material, which contains or is contaminated with radionuclides at concentrations or radioactivity levels greater than the clearance level as established by the regulatory body without foreseen further use. In China, RAW arises principally from NPP, research reactors, the nuclear fuel cycle, nuclear technology applications, the exploitation and utilization of uranium and thorium resources, as well as clean-up activities of contaminated sites and/or facilities such as that shown in Fig. 22.2: some nuclear facilities in the Gobi Desert in the west part of China (Qinghai Province), which were used during the 1950s and 1960s, need to be cleaned up.

To meet the needs for its nuclear power expansion, China has developed uranium enrichment and fuel element manufacture capability. At present, two uranium enrichment plants are in operation, with annual total centrifugal enrichment capacity of 1,100 tons of separation work. The first nuclear fuel assembly production line was established in 1988 in Sichuan province, supplying most of the nuclear fuel elements to the Qinshan NPP (Fig. 22.3). Subsequently, the technologies for designing and manufacturing nuclear fuel elements have been imported on a step-by-step basis, to which a technical adaptation was later made. This means that China's PWR fuel element manufacture can meet the requirements of the international generic standards, so as to ensure that the supply of nuclear fuel elements meets the demands of the current PWR plants in China. Through introducing technology from Canada, a high pressure reactor fuel element production line, with

22.2 Nuclear facilities in the Gobi Desert in Qinghai Province in the west part of China, which were used in the 1950s and 1960s, need to be cleaned up.



22.3 Qinshan nuclear power plant with five reactor units.

a capacity of 200 tonnes per year, was built in Inner Mongolia, Northern China, where it provides HWR fuel elements for Qinshan NPP III.

China's RAW categorization system is based on pre-disposal management and disposal of RAW. In pre-disposal management, the RAW categorization system accounts for the nuclear facility operational experience in waste treatment and conditioning requirements, which includes a

quantitative categorization system for radioactive gaseous, liquid and solid wastes. The disposal-based RAW categorization system focuses on the final disposal of RAW, in conjunction with the origin of the waste and the planned disposal approach.

The pre-disposal management-based waste categorization system is used to manage gaseous, liquid and solid RAW generated at nuclear facilities, with a detailed categorization for different forms of wastes according to their radioactive characteristics as shown in Table 22.3. This is consistent with the basic requirements of waste treatment but puts more emphasis on

Table 22.3 Pre-disposal-based waste categorization system

Physical condition	Waste categorization	Waste characteristics/index
Gaseous	Low level waste (LLW)	Concentrations not exceeding 4×10^7 Bq/m^3
	Intermediate level waste (ILW)	Concentrations greater than 4×10^7 Bq/m^3
Liquid	Low level waste (LLW)	Concentrations not exceeding 4×10^6 Bq/L
	Intermediate level waste (ILW)	Concentrations greater than 4×10^6 Bq/L, but not exceeding 4×10^{10} Bq/L
	High level waste (HLW)	Concentrations greater than 4×10^{10} Bq/L
Solid	Low level waste (LLW)	Specific activity not exceeding 4×10^6 Bq/kg.
	Intermediate level waste (ILW)	1. Half-life longer than 60 days but shorter than or equal to 5 years, specific activity not exceeding 4×10^6 Bq/kg 2. Half-life longer than 5 years, but shorter than or equal to 30.2 years, with specific activity more than 4×10^6 Bq/kg, but not exceeding 4×10^{11} Bq/kg 3. Half-life longer than 30.2 years, specific activity greater than 4×10^6 Bq/kg.
	High level waste (HLW)	1. Half-life longer than 5 years, but shorter than or equal to 30.2 years, with heat release rate larger than 2 kW/m^3 or specific activity more than 4×10^{11} Bq/kg, 2. Half-life longer than 30.2 years, specific activity larger than 4×10^{10} Bq/kg.
	Alpha radioactive waste	Alpha nuclides with half-life longer than 30.2 years, specific activity in a single container larger than 4×10^6 Bq/kg

the cleaning index, shielding design, and other field protection requirements. These requirements are implemented in the waste treatment and conditioning processes for various systems. It is noticeable that most Chinese standards on nuclear or radioactive waste management are coherent with the current IAEA classification scheme. For example, both the IAEA and Chinese standards specify that management of decay heat should be considered if the thermal power of waste packages reaches several watts per cubic metre [10,11].

The disposal-based radioactive waste categorization system divides solid radioactive waste into solid LLW, solid ILW, solid HLW, solid alpha waste and the waste arising from mining and milling of uranium and thorium, and naturally occurring radioactive materials (NORM) waste. Disposal options considered include centralized deep geological disposal, regional near-surface disposal, and centralized landfill, and others, as shown in Table 22.4. Solid LLW containing only short-lived radionuclides can be released from regulatory control when the radioactivity contained is below the regulatory clearance levels. However, management of cleared waste should be in compliance with other relevant environmental requirements.

22.2.1 Low and intermediate level radioactive waste

Low and intermediate level radioactive waste (LILW) arises mainly from NPP operation and nuclear technology applications. Radioactive waste produced from operating NPPs is principally from the following:

- main process equipment and waste treatment equipment, including secondary waste from loop leakage or drainage and waste treatment systems, which includes airborne and liquid radioactive wastes,
- technical maintenance during operation,
- protective articles such as shielding, equipment and miscellaneous scrap replaced during the daily operation.

Table 22.4 Disposal-based radioactive waste categorization system

Waste category	Disposal approach
Solid HLW	Centralized disposal
Solid α waste	Centralized disposal
Solid ILW	Regional near-surface disposal
Solid LLW	Regional near-surface disposal
Uranium (thorium) mining and milling waste	Backfilling, damming, centralized landfill
Naturally occurring radioactive materials (NORM) waste	Backfilling, damming, centralized landfill

The wastes arising from nuclear technology applications refers to contaminants that arise from the applications of radioisotopes and irradiation technology in industry, agriculture, medicine, research and teaching, which contain:

- man-made radionuclides with specific activity higher than 2×10^4 Bq/kg;
- or NORM wastes with specific activity higher than 7.4×10^4 Bq/kg;
- or abandoned/discarded wastes arising from the above-mentioned activities with surface contamination levels exceeding the regulatory limits.

Such LILW is widely distributed, of a wide variety, and usually in small amounts.

22.2.2 High level radioactive waste

HLW includes the high-level liquid waste generated from the reprocessing of SF, and the solidified form of such waste, as well as SF withdrawn from reactors or research reactors pending direct disposal. Due to its high activity, large heat release, high toxicity and long half-life, HLW needs to be isolated from the human environment for a long period of time in a reliable manner.

Uranium (thorium) mining and milling waste

Uranium (thorium) mining and milling wastes have radioactivity levels exceeding the relevant regulatory levels. They were generated from exploration, mining, milling closure, mainly covering barren rocks, and tailings characterized by large volume, low activity and simple radionuclide composition.

Naturally-occurring radioactive materials (NORM)

NORM means wastes containing, or contaminated, with naturally occurring materials at a concentration or radioactivity higher than the relevant regulatory level and which is expected to have no further use. These wastes arise principally from the mining and milling of rare-earth minerals and the production of phosphates among others. The radioactivity in such kinds of wastes is mainly from radioactive materials associated with raw materials and of quite large volume.

Spent fuel (SF)

The amount of Chinese SF was about 1,000 t from light reactors in 2010. It will be 2,000 t in 2015 and then 1,000 t produced each year from 2015 to

2020. However, a single CANDU reactor which will be in operation in Qinshan III will give 200 t SF each year when it is in operation. Since 2010, SF from China's LWRs is being reprocessed first in a small pilot plant, followed by vitrification and eventually geological disposal.

22.3 Radioactive waste (RAW) management strategies: history and developments

China adheres to the RAW management policy of educating the people, developing harmony between the people and the industry, making accident prevention a priority, adhering to strict management and a high safety priority: overall a combination of prevention and management. RAW is managed by taking all reasonable and practicable management measures in such a way that it will not impose undue burdens of waste management on future generations. Radioactive management ensures adequate protection of humans and the environment now and in the future to ensure the sustainable development of beneficial human practices. Both the RAW management legislative and regulatory systems and the independent RAW safety regulatory systems are established in China. A licensing system for RAW management activities is currently in operation and the licence holder is responsible for the safety of RAW and relevant management facilities. Through reasonable selection processes for the raw materials, use of advanced production processes and equipment, implementation of re-use and recycling when possible, it is anticipated that the current practices can significantly reduce the generation of RAW as well as the release of any radionuclides to the environment, to a level as low as reasonably achievable (ALARA).

Simultaneously, the relevant RAW treatment facilities have been established with the main technological process in their design, construction and operations. The licence holders have to solidify liquid RAW in a timely manner and limit the storage duration for both liquid waste and solid waste. RAW management in China is oriented to proper disposal and discharge and both are implemented over the whole spectrum of waste streams and for the whole process from cradle to grave. Solid RAW is disposed of in accordance with its categories; uranium mining and milling wastes are disposed *in situ*; and solid LILW will be disposed of in near-surface disposal facilities. Solid HLW and alpha wastes are planned to be disposed of in a centralized deep geological disposal repository. Research and development into HLW geological disposal is performed through strategic planning, harmonized development, step-wise decision making, and is an iterative process.

22.3.1 Treatments and conditioning of radioactive wastes

With the rapid development of its nuclear industry, China's RAW management has gradually been improved over the past 20 years. In the 1950s, when the country's nuclear industry had just begun to develop, the Chinese government put forward the policy that radiation protection should be developed before the nuclear industry became operational, which required that any work involving radioactivity must be accompanied by waste treatment capability and that any RAW discharge complies with the required standards. Therefore, nuclear industry production and research facilities were all equipped with RAW treatment and storage installations for storage of different categories of wastes in accordance with the categorization given in Table 22.4.

In the early years, the liquid and gaseous radioactive waste treatment processes, as part of nuclear production and research activities and as a component associated with the main production process, employed purification filtration, evaporation, and ion exchange among other practices. Such wastes were discharged into the atmosphere and surface water after meeting the national standards – 'Radiation Protection Regulations' (GBJ8-74) [11,12]. This standard was issued by the State of Ministry of Nuclear Industry, targeting the national regulations on the treatment and disposal of radioactive wastes. Those liquid and solid radioactive wastes that could not be discharged were stored. In general, in the process of nuclear facility construction and operation, the treatment of gaseous and liquid radioactive waste generally received due attention with practical treatment technology being employed. This played an important role in ensuring normal operation as well as environmental protection. All sorts of liquid wastes generated in the processes operating at each nuclear facility underwent solidification treatment. Evaporator residues of liquid LLW underwent bituminization and the resultant solidified forms, after packaging, were sent to a storage facility near Beijing. The programme for dealing with China's legacy HLW is based on joule heated ceramic melters (JHCM) such as those used in Germany, Japan, the US and Russia operating at well over 1,100°C. However, opportunities exist in the future that waste streams from NPP from China may be more applicable to cold crucible induction melting (CCIM) technology, which has been developed intensively by France and Russia. From a materials point of view, selected glass compositions will be within the borosilicate range adapted for current wastes and the envisaged future HLW streams. Large-scale research programmes and investment are also under way on the development of glass composite and ceramic waste forms.

With the construction and expansion of NPPs and the development of the radioactive waste management concepts of making safe disposal central,

progress has been made in RAW treatment and conditioning technology and installation. NPPs in China now have liquid and solid RAW treatment facilities installed during their construction. NPP operators prepare RAW management programmes, which specify the assignment of responsibility for RAW management within each NPP. The Chief Manager of each nuclear operational organization acts as the primary person responsible for RAW management. The Chief Manager is responsible for providing sufficient resources to ensure effective implementation of the RAW management programme, and to ensure the national limits of radioactive effluents are complied with. This RAW management arrangement can be maintained and modified in a sustainable manner.

RAWs are managed according to their categories at NPPs. Based on the features of each NPP, the specific categorization schemes are developed and applied to the management of RAW arising from NPP operations. In general, concentrated liquid and spent ion exchange resins are solidified in cement, the waste arising from technology processes is held in storage after sorting and compression. Cement solidification processes have been established in Daya Bay, QNPP II and Ling-ao NPPs to carry out cement solidification of liquid LILW, spent exchange resins and spent filter cartridges. Spent ion exchange resins produced at QNPP and QNPP III are currently stored temporarily and cemented waste forms are stored in waste storage facilities at such NPPs. The solid RAW generated at NPPs is mainly stored in on-site facilities and the liquid wastes are stored in tanks. On the whole, the facilities for waste storage at NPPs are well constructed and in a good condition, and comply with current requirements.

In China, the NPP operators continue to carry out technology modifications. QNPP upgraded the cement solidification installation and as a result the waste drum-filling coefficient increased from less than 79% to more than 90%. Guangdong Daya Bay NPP continues testing to improve the formula for cementation of its spent ion exchange resin so as to raise the waste loading capacity. Daily operational practices include measures to control waste generation. Personnel awareness of waste minimization is reinforced through training and education activities. Suitable operational processes are employed and technological and administrative measures are envisaged to make waste generation ALARA. Moreover, detailed work plans and arrangements to control waste generation during maintenance include:

- control of waste transfers to prevent contamination,
- maintenance of normal operations of the waste treatment system to reduce generation of secondary waste,
- minimization of the entry of materials into controlled areas, and

- enhanced recovery and re-use by dismantling the disused intermediate and high efficiency filters, and returning metal frameworks to manufacturers when the contamination is below clearance levels.

As of December 2006, the volume of solid LILW generated from China's NPPs was 4773 m^3.

Tracking solid RAW is an important aspect in its safety RAW management. Each NPP writes specific management procedures to require the tracking of its RAW. Each waste package is tracked by establishing a unique RAW record. The relevant information of the record includes origin of waste, type of waste, date of waste generation, radioactivity level in waste, quantity/volume of waste, temporary storage location, etc.

A main objective of RAW management is to minimize the generation of RAW in China. Compared with some countries, there is still potential to reduce waste generated at China's NPPs. However, the minimization of RAW is a combined effort balancing factors of technology, safety and economy. China is taking additional actions in controlling the generation of the wastes, upgrading management practices, introducing advanced waste reduction technologies, promoting specialization and socialization in RAW treatment services.

22.3.2 Sealed spent radioactive sources

Sealed spent radioactive sources are currently held in the provincial nuclear waste storage facilities, in the centralized sealed source storage facility, or at the user's premises. These radioactive sources have not been conditioned into a stable form, so they occupy large volumes of storage space and pose high potential risk. China is making an effort to establish an R&D base to develop radioactive source conditioning technology as soon as possible for the purpose of improving the safety of radioactive source storage. At the same time, China is exploring options for disposal of spent radioactive sources; it is expected to seek a long-term solution for spent radioactive sources. To meet the need for application of radioactive sources, since the 1960s China has invested in constructing a different scale of storage facilities in Beijing, Changchun, Lanzhou and Wuxi to accept and store RAW arising from nuclear technology applications, including disused sealed sources.

22.3.3 Radioactive waste storage in China

To keep pace with the development of nuclear technology applications, temporary storage facilities have been constructed in China since the 1960s. The *Notification on Strengthening Radioactive Environment Management*

Arrangement was issued in the *Temporary Regulations on Construction of Urban Radioactive Waste Repository* in 1984. The Methods *on Urban Radioactive Waste Management* was issued in 1987 [12,13]. Temporary waste storage facilities are constructed on a provincial basis. Each province (or autonomous region, or municipality directly under central government) builds one such facility to accommodate wastes arising from research, teaching, medicine and other applications of radioisotope and nuclear technology within the province. Provincial environmental protection agencies have set up special organizations staffed with specialists responsible for supervision and environmental monitoring. The *Criteria on Siting, Design and Construction of Application Waste Storage Facility* was issued in 2004 [14] and requires the modification and extension to be carried out for existing storage facilities to meet the new requirements. At present, special funds have been appropriated for this purpose. It also requires an environmental impact assessment to be made prior to such modification and extension, which cannot be implemented without approval by the relevant agencies. By the end of 2010, a total of 31 waste storage facilities, together with one centralized storage facility for spent radioactive sources, had been constructed and/or upgraded in compliance with the new requirements. At the end of 2006, these facilities had received 64,572 m^3 of disused sealed sources, of which 49,741 m^3 are in the provincial storage facilities, and the remainder is in the national centralized facility.

22.3.4 Low- and intermediate-level radioactive waste disposal

In the 1980s, radioactive waste disposal work was initiated in China. The former Ministry of Nuclear Industry (MNI) subsidiary Science and Technology Committee set up a panel to examine RAW treatment and disposal. The siting of solid LILW disposal facilities began in the 1980s and was implemented under the auspices of the former MNI. The initial siting work was conducted in South China, East China, Northwest China, and Southeast China based on the distribution of nuclear facilities at that time. Determination of the South China disposal site began in 1991, with 27 candidate areas being selected. Of these, 20 were investigated on site and three candidate sites were identified. In 1998, initial reconnaissance was carried out within the area of Zhejiang province, East China, with 17 areas surveyed and five candidate sites identified. In Northwest China, two candidate sites were identified on the basis of six surveyed areas. After further comparison, a disposal site in the northwest was determined. In southwest China, examination of disposal sites was carried out from 1989 to 1991. The site survey

was carried out in ten candidate areas selected from an initial 38 areas, of which three candidate sites were finally recommended.

China's Environmental Policy on Disposal of LILWs was issued in 1992 (hereinafter referred to as Paper 45) [15], which clarify the environmental policy on LILW. Paper 45 states that national disposal facilities for LILWs shall be constructed in the regions where major waste generation occurs in order to dispose of LILWs generated in the region and neighbouring regions. Paper 45 played an active role in promoting the siting and construction of LILW disposal sites. In 1998, construction of the Northwest disposal facility was completed, with planned capacity of 200,000 m^3. The first phase of construction was planned to generate 60,000 m^3 of disposal capacity, and so far 20,000 m^3 has been constructed. The Northwest disposal facility is currently in trial operation. By the end of 2006, this site received 471 m^3 of LILW with total activity of 3.05×10^{12} Bq. In August 2000, Guangdong Beilong, China's second solid LILW disposal facility was constructed in the Guangdong Province with planned long-term capacity of 240,000 and planned near-term capacity of 80,000 m^3. The total capacity that has been constructed in the first phase was about 8,800 m^3 and, by the end of 2006, the received waste amounted to 1403.2 m^3. Environmental monitoring indicates that operation of these two LILW disposal sites has no negative impact on the surrounding environmental radiological levels and no radiation accident has occurred to date.

Under the Law of the People's Republic of China on Prevention and Control of Radioactive Pollution of 2003 [9], the relevant government agencies are developing the national programme of finding solid radioactive waste disposal sites. The principle is to make an overall plan and implement the project in a step-wise, convenient and economical way to ensure safety. Based on the future development of NPPs and the distribution of waste generation varying with time and region, the overall development programme for LILW disposal will be established including allocation of regions, siting planning, capacity of disposal site and construction plan. Based on the programme, a phased implementation approach shall be developed to keep the number and capacity of disposal sites countrywide adequate to meet the demand for RAW disposal in the various regions. Construction of disposal facilities on the sites that have been chosen should be implemented in phases based on the quantity of LILWs generated and on a basis of gradual disposal capacity extension so as to achieve the effective disposal capacity. When considering the safety of LILW disposal, transportation is one of the most important factors. Full account must be taken of the safety, economics, and convenience of RAW transport. To this end, a reasonable arrangement should be made for the coverage of each regional disposal site.

22.3.5 Post-closure of disposal facilities

Requirements on surveillance control of disposal facilities after closure have been laid down in China. The Regulations on Radioactive Waste Safety (HAF401) require that, after closure of a disposal site, institutional surveillance and control should be maintained to:

- prevent inadvertent public intrusion onto the site,
- prevent movement and disturbance of disposed radioactive materials,
- monitor the performance of the disposal site against design basis standards, and
- implement necessary remedial actions.

The period following closure of a disposal facility generally includes closed, semi-closed and open phases. Closed phase means a period when the disposal facility that has just been closed is kept under closed condition and that no one can access it unless for the purposes of a supervisory task. Semi-closed phase means a period when waste is covered with well-structured cover and associated hazards has proven very small, and people are allowed access but without any activities relevant to drilling and excavations. Open phase means a period when radioactivity of waste has reduced to the level at which radiation protection is no longer needed following expiration of the required control period and the site can be fully open to the outside.

Post-closure surveillance of the localities where a disposal facility is located are the duty of the local government. Costs required for carrying out post-closure maintenance, monitoring and emergency measures are estimated before the operation of such a disposal facility and collected in an appropriate amount from the associated waste disposal fees. Re-estimation, and necessary adjustment, can be made for such costs to meet the changing circumstances. Post-closure supervision, such as environmental monitoring, access restriction, installation maintenance, file preservation and possible emergency actions, should be carried out under the auspices of the environmental protection agencies at both the national and the provincial levels. Both the Guangdong Beilong and the Northwest China LILW disposal sites are in operation, and far from closure.

22.4 Geological disposal of high level waste (HLW)

Under the Law of the People's Republic of China on Prevention and Control of Radioactive Pollution [9], the competent nuclear facility authority under the State Council (National Atomic Energy Agency), in conjunction with environmental competent authority under the State Council, has been developing a programme for the siting of a HLW geological disposal

facility (GDF). The siting programme is based on the geological conditions, the solid RAW disposal needs, and the associated need for an environmental impact assessment. This programme cannot be implemented without the approval of the State Council. Based on such a programme, the relevant local governments would provide construction land for the solid RAW repository and take effective steps to support the disposal of such waste. Disposal of solid RAW on any inland river and or marine environment is prohibited. The Law of the People's Republic of China on Prevention and Control of Radioactive Pollution defines that HLW shall be disposed of in a centralized deep geological repository.

The 'Guidance on Siting of Radioactive Waste Geological Repository' states that the basic aim is to select a site suitable for disposal of HLW, where the disposal facility and waste package would be able to effectively isolate radionuclides from entering into the biosphere over geological timescales. The site could provide one or more natural barriers to keep the adverse impacts on the population and the environment at the acceptable level specified by the national regulatory body. Studies of deep geological disposal of HLW in China began in 1985, when the initial R&D programme was initiated under the auspices of the former MNI in respect of engineering, geological, chemical and safety issues. Experimental facilities were established to simulate the chemical environment of potential geological disposal environments. At the same time, a preliminary safety assessment of geological disposal was launched. A study on the pre-siting of HLW disposal facility was also conducted. Preliminary regional comparisons have been performed for five regions: East China, South China, Southeast China, Inner Mongolia and Northwest China. However, the characterization work has focused on Northwest China.

In 2006, the Guides on Research and Development Planning of Geological Disposal of HLW were issued jointly by China Atomic Energy Administration (CAEA), Ministry of Science and Technology (MOST), and China's regulator the State Environmental Protection Administration (SEPA). The overall purpose of the study on geological disposal of HLW in China is to select the potential site with stable geological and suitable socio-economic environment and then to complete the construction of the country's geological disposal facility for solid HLW in a manner that protects the environment and the public from unacceptable hazards through the containment and retardation effects of engineered and geological barriers.

Under these guidelines, geological disposal of HLW R&D is divided into three stages:

1. laboratory R&D and siting of the disposal facility (2006–2020),
2. underground experimentation (2021–2040), and

3. demonstration of a prototype disposal facility and demonstration and construction of such disposal facility (2041–mid-21st century).

Around 2020, the following tasks are expected to be completed:

- the in-laboratory R&D project involving multidisciplinary fields,
- preliminary siting of a disposal facility,
- a feasibility study for an underground laboratory, and a safety review for construction of an underground laboratory.

Around 2040, the following tasks will be completed:

- R&D for the underground laboratory,
- preliminary confirmation of the disposal facility site,
- pre-feasibility study report of disposal facility, and
- feasibility study and safety review of prototype disposal facility.

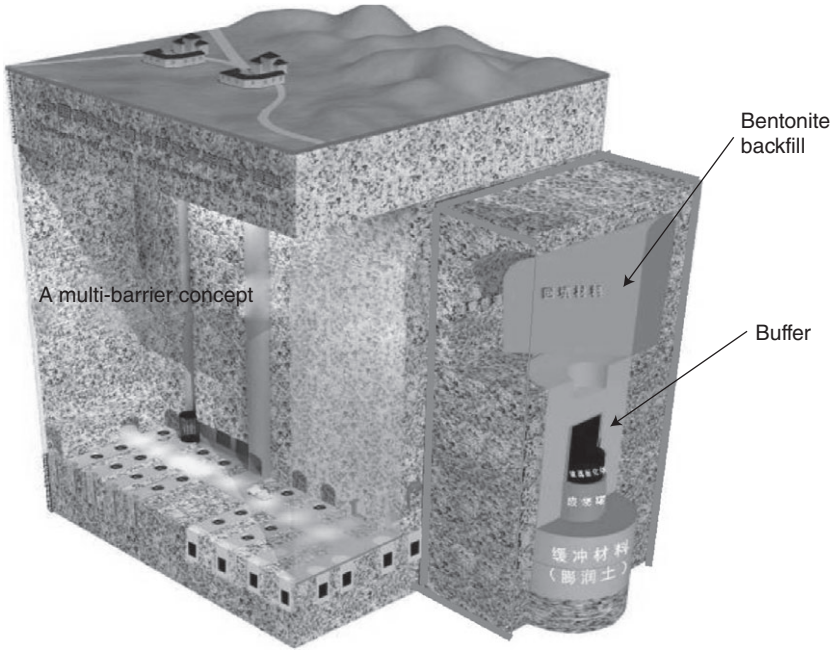
From 2040 to the mid-twenty-first century, the following objectives would be achieved:

1. demonstration experiments of the prototype disposal facility,
2. final confirmation of disposal facility site,
3. feasibility study of the disposal facility and safety assessment for the disposal facility construction,
4. disposal facility construction, and
5. the safety review for disposal facility operation.

22.4.1 China's HLW/SF repository concept

Public concerns over the global ability to manage, and eventually dispose of RAW, especially HLW/SF, remain high. Emplacement in the deep geology is an internationally recognized disposal solution for HLW and SF, and China is planning to use this route. While China's GDF programme is at an early stage, like all international waste management programmes implementing geological disposal, it is considering multi-barrier concepts comprising engineered and natural barriers between the HLW/SF in the geosphere and the biosphere, while bentonite-based engineered barrier systems (EBS) were considered in China as early as the 1990s [16–17]. The current preliminary geological disposal concept for its HLW/SF is to use a shaft-tunnel model in the saturated zones of granite rock (Fig. 22.4). Over the past 20 years, China has made great strides in its geological repository programme including, as described above, geologically surveying the whole country for its georepository site selection and optimization of backfill/buffer materials that will be needed for the GDF safety cases [18].

Many other countries are developing similar concepts for permanent disposal of radioactive waste deep underground: solidification of HLW/SF



22.4 China's preliminary HLW repository concept.

using glass and ceramics, packaging in metal canisters, following temporary storage above ground before permanent geological disposal in natural barrier systems such as a granite rock-body, using a multi-barrier system [16,17]. Chinese researchers have suggested that EBS is a major component in guaranteeing long-term safety, making it necessary to conduct fundamental research on the coupled THMC (thermal-hydrological-mechanical-chemical) behaviour of bentonite under simulated geological disposal conditions, and subsequently to reveal the property changes of the bentonite over a long period of time.

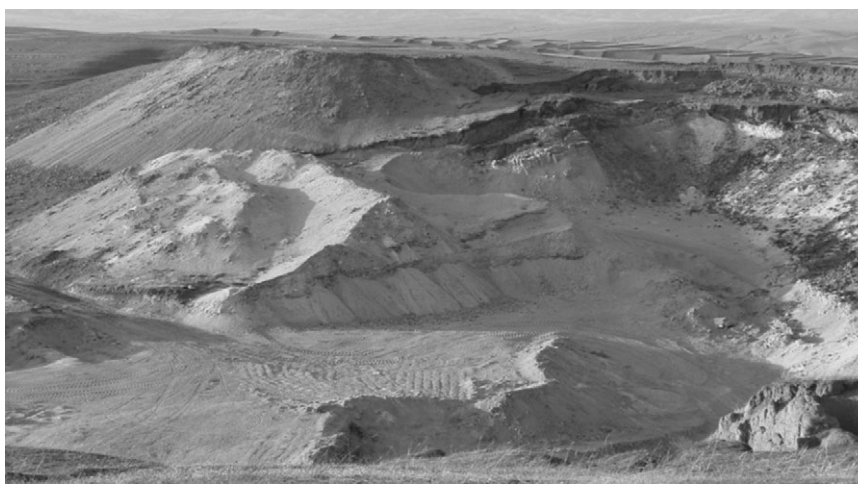
The requirements for HLW backfill materials are long-term chemical and physical stability, good mechanical properties, volume expandability in contact with water and very low water penetrability. Other requirements also include the ability to hinder nuclide migration, good thermal conductivity and thermal stability, radiation resistance and stability, natural availability and importantly, low cost.

Many years of research in Europe and China on bentonite backfill materials for the EBS has revealed that bentonite comprising predominantly montmorillonite is considered to give the best performance in terms of low water penetration, high volume expansion, and excellent nuclide absorption and retention, as well as being abundant.

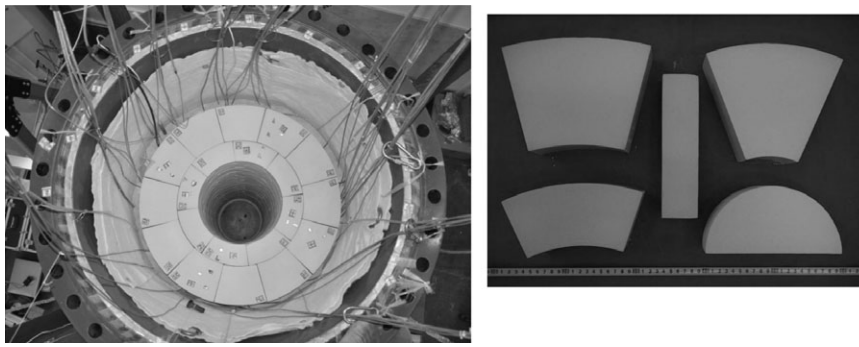
China is rich in mineral reserves and has large bentonite reserves suitable for the EBS backfill/buffer (at one site with a volume of $40 \times 40 \times 0.7$ km) in China's Inner Mongolia region near Beijing. Bentonite with high content of expandable montmorillonite has been found in an area named Gao-Miao-Zi (GMZ, which in English means Highland Temple). This single reserve, as shown in Fig. 22.5 is over 280×10^6 tonnes.

It is expected that the bentonite at Gao-Miao-Zi will be used in China's HLW/SF geological repositories. This bentonite is being considered as a part of the EBS due to its ability to retain radionuclides and other hazardous materials. Prior to considering modular designs for canister encapsulation in the GDF, bentonite natural resources, raw mineral analysis, characterization and processing, need to be investigated, developed and optimized for large-scale cost-effective manufacture. To demonstrate the long-term safety of a GDF in China, the influence of the bentonite composition and the properties of the compacted block/brick must be studied.

Some large-scale mock-up facilities have also been built in China to test the efficacy of backfill/buffer materials such as bentonite with designed canisters. A China mock-up test was recently initiated after a long period of research conducted with international support. It is based on a preliminary concept of the HLW granite rock environment repository in China [19]. It was developed to investigate the THMC properties of compacted GMZ-Na-bentonite as shown in Fig. 22.6, which reveals the arrangement of compacted bentonite mineral blocks inside the mock-up test steel. The work has been carried out and led by the Beijing Research Institute of Uranium Geology (BRIUG) [20–22]. The device contains a heater, which



22.5 China bentonite GMZ 1,600 million tonnes of Na-based bentonite site in Gao-Miao-Zi, Inner Mongolia, China.



22.6 Compacted engineered bentonite blocks arranged in inside a mock-up test facility within a steel tank (top view). This mock-up THMC test consists of a heater (canister) and bentonite blocks within a cylindrical steel tank.

simulates the heat from a container of HLW/SF, placed inside the compacted GMZ Na-bentonite blocks with total dry density 1,600 kg/m^3. Water inflow through the barrier from its outer surface simulates the water penetration. The device is a large steel tank of 900 mm internal diameter and 2200 mm in height. The experiment is being performed at the BRIUG laboratory and the design concept is shown in Fig. 22.6 and Plate VI (between pages 448 and 449). In Fig. 22.6, the compacted engineered bentonite blocks are arranged inside a mock-up test facility within a steel tank (top view). This mock-up THMC test consists of a heater (canister), bentonite blocks within a cylindrical steel tank, as shown in Plate VI as a sketch of the cross section of the China mock-up facility and the arrangement of central heater, steel canister, bentonite blocks/bricks and multiple sensor arrangement [22].

22.4.2 Current status of China's repository programme

After more than 20 years of geological survey and investigation, the Beishan area was chosen as one of China's likely areas for its GDF for HLW/SF. Beishan, in the Gobi desert, is extremely dry and has been unchanged for millions of years. It is located in a remote area of Gansu Province in northwest China, not far from the west end of China's Great Wall, Jia-Yu Guan (Jia-Yu Fortress). The narrow aubergine-like Gansu province is also referred to as Western River Corridor (Yellow River West Corridor), linking central China to China's Xinjiang Autonomous Region – crossing and along the Gobi Desert through about a thousand miles linking to the west part of Asia and Eastern Europe as shown in Plate VII (between pages 448 and 449).

A thorough geological survey has been carried out at Beishan (Fig. 22.7). In August 2005, the CAEA revised the long-term HLW geological repository programme, with the objective of building China's HLW geological repository by about 2050. China is closely monitoring the potential environmental impacts of nuclear energy for future generations, particularly where HLW/SF and geological disposal are concerned. China's regulator body, the SEPA, implements the activities related to radioactive waste and disposal, which have been managed by the China National Nuclear Corporation (CNNC). Furthermore, China's HLW/SF geological disposal R&D programmes are carried out by CNNC's research and engineering organization and led by BRIUG.

The Chinese government has approved a three-phase GDF programme:

1. Phase I: Site selection and site confirmation (2001–2020): Technical preparation, HLW disposal/repository programme started in China; geological study, preliminary site characterization and evaluation: investigations on surface geology, hydrogeology and geophysics with the drilling of four boreholes (BS01-04) and *in-situ* tests in boreholes.
2. Phase II: Underground Research Laboratory (URL) construction and *in-situ* tests (2010–2030): *in-situ* tests on EBS on backfill/buffer materials, radionuclide migration and use of the necessary natural analogues; mock-up tests and underground lab tests of backfill/buffer materials, together with coupled THMC tests.
3. Phase III: Repository construction (2030–2040): Construction structural design, simulation and modelling, construction and preparation of geological repository engineering work.

While the granite site at Beishan is regarded as the most likely site, China is keeping its options open by also examining a potential GDF site in clay formations in Xinjiang in northwest China.

22.7 Beishan area is part of Gobi Desert (left). Beishan drilling site while engineering drilling was in operation (right).

22.4.3 Progress at the Beishan site

Beishan is regarded as the most likely area for China's GDF, because there is/are:

- no economic prospects for the Gobi desert area, future possibilities may be in wind energy and solar energy but they will be located at the surface;
- very low population density and the prediction is that it will remain the same in the foreseeable future since there are no important mineral resources;
- extremely low rain fall (60–80 mm/year) and very high evaporation rate (2900–3200 mm/year),
- convenient transportation, as it is on the edge of the Gobi desert and about 200 km from the main east–west train lines and motorways.
- favourable geology with stable granite and diorite rocks and suitable hydrogeological conditions;
- international programmes in similar granite host rock using the multi-barrier concept [16–18].

Progress at Beishan includes site selection of an area covering hundreds of square miles with a crust thickness of 47–50 km and no earthquakes with magnitudes over 4.75 on the Richter scale ever having taken place. The topography of the area is flat with some small hills with elevations above sea level ranging from 100–2,000 m. Original site characterization, in particular its hydrogeological properties, showed very poor groundwater resources. Average precipitation is 70 mm/year while evaporation is about 3,000 mm/year and there is no year-long stream and other surface water body in the area. Geo-stress and borehole measurements also gave positive results with tensile strength from 5 to 7 MPa and compression strength from 5 to 13 MPa, reflecting samples obtained from a depth of 200–500 m, while maximum lateral stress reached is 25 MPa at a depth of 500 m.

22.4.4 Public involvement in the siting process

The Law of the People's Republic of China on Environmental Impact Assessment states that for projects that may have adverse environmental impact, public meetings should be held or other approaches adopted to solicit comments on the draft environmental impact assessment statement from relevant organizations, experts and the public before its submission for review and approval. The constructor and the operator of the proposed site will need to take into consideration the comments provided from the relevant organizations, experts and the public, and provide additional

explanations on whether these comments have been incorporated when submitting the environmental impact assessment report for review.

22.5 Future trends

China's strategy for RAW management is to minimize waste volume, to reduce large volume in processed wastes, and to dispose of LILW in near-surface disposal facilities. Meanwhile, the vitrifying of high level waste (HLW) has to be carried out and the vitrified HLW has to be disposed in a geological repository facility.

The FBR development programme in China has a three-stage strategy beginning with an experimental fast reactor (CEFR) at CIAE to develop fast reactor technology with a sodium-cooled pool-type reactor with 65 MW (25 MWe) capacity. While China aims eventually to develop its own capability in all aspects of the fuel cycle, it recognizes that in the meantime it will need to draw on international expertise to gain key technical skills in reactor design, construction, and operation and particularly in NPP decommissioning and HLW waste disposal.

To underpin its nuclear plans, China is already participating in a number of international programmes including training and education exchange programmes in Europe, Generation IV reactor development, and research into partitioning and transmutation. International joint research programmes in geological disposal and repository are carried out in China in the areas of coupled THMC for backfill/buffer materials, mock-up and URL, repository site selection and natural analogues, in collaboration with various international bodies, including IAEA, CAD/CEA, POSIVA, SCK-CEN, SKB, NAGRA, JAEA/RWMIC, KAERI, CUT and BGR/DBE/GRS.

However, key scientific challenges remain for China's waste management programme. Nuclides in HLW/SF disposal are a scientific, technological and a social challenge due to the fact that their safe and reliable disposal in repository has to be fully isolated for hundreds of thousands of years, possibly millions of years due to these highly radioactive nuclides with extremely long half-life (i.e., Np, Pu, Am, Tc, etc.). These determined the complexity and difficulty in terms of site selection, the EBS evaluation, deep repository design, construction and long-term safety assessment. To resolve these challenging issues, a great deal of investment is needed for carrying out the following large-scale R&D projects:

- developing and testing excavation technologies,
- establishing the excavation damage zones,
- site characterization,
- hydrogeological tests,

- *in-situ* radionuclides migrations tests,
- simulation of effects caused by emplacement of RAW,
- demonstration of EBS,
- prototype repository study,
- natural analogue and anthropogenic analogue studies (i.e., the Lianshan-guan uranium deposit study in China).

Meanwhile, only limited knowledge has been obtained on the geochemical process of the radionuclides under the natural disposal/repository conditions. Other challenges include nuclides like T_c, ^{131}I and ^{3}H, which are difficult to immobilize or retard once mobile.

It is considered that a long period of time will be needed for the safety assessment of a GDF, which must cover wasteforms, canisters, buffer materials, near, and far field, biosphere and geosphere, and most importantly the groundwater systems. Research developments have to involve geology, hydrogeology, radiochemistry, rock mechanics, engineering, material science, mineralogy, with coupled THMC EBS behaviour, and overall radiation protection technology, policy and regulation bodies. Key challenges that need to be solved in China's GDF programme are:

- reliable predictive modelling of the evolution of a repository site,
- characterization of deep geological environment,
- behaviour of deep rock mass, groundwater and engineering materials under coupled conditions up to high temperature,
- *in-situ* stress, hydraulic, chemical, biological and radiation processes,
- geochemical behaviour of transuranic radionuclides with low concentration and their migration in groundwater,
- safety assessment of disposal system,
- regulation and policy associated with the HLW/SF GDF.

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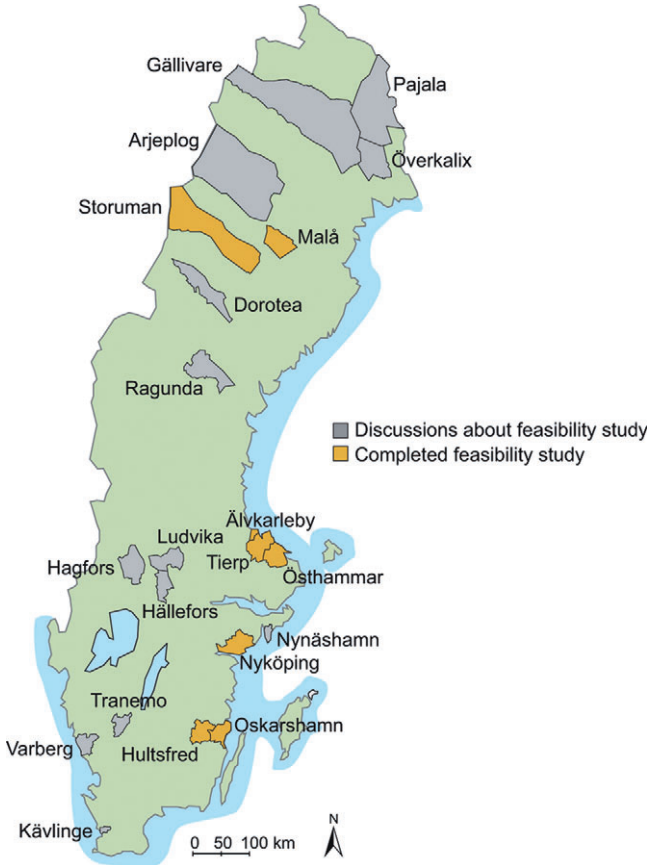

Plate IV (Chapter 13) Counties where SKB considered feasibility studies (from SKB Report R-11-07).

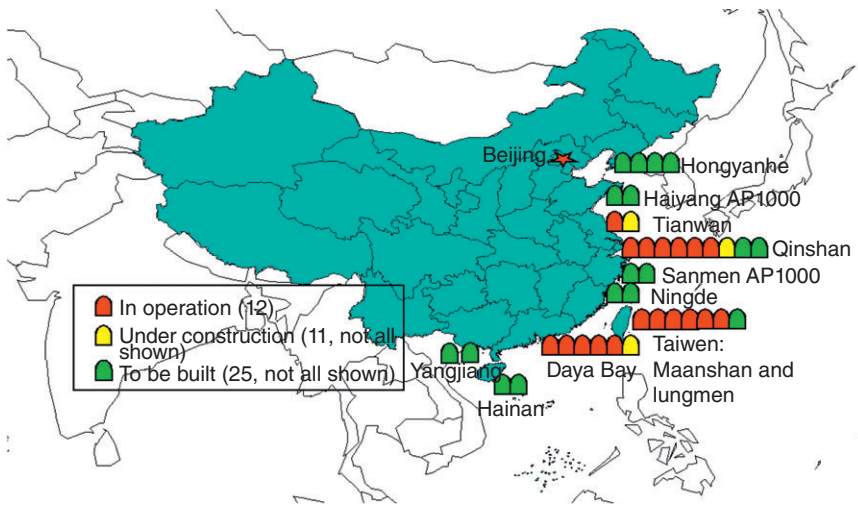

Plate V (Chapter 22) Distribution of NPPs in China.

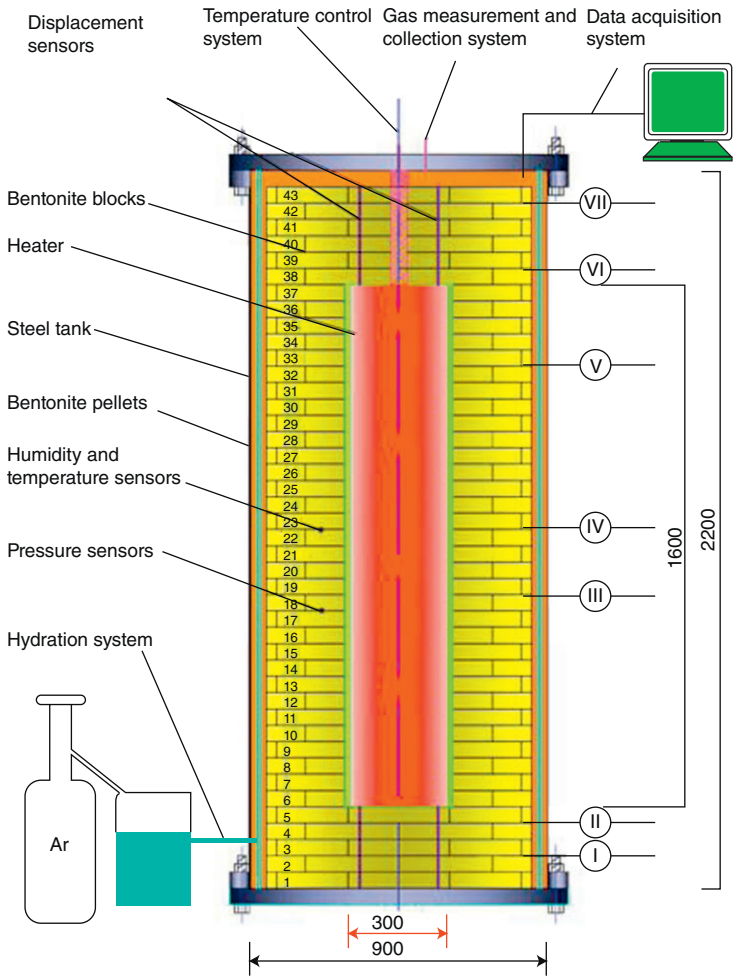

Plate VI (Chapter 22) Sketch of the cross-section of mock-up facility and the arrangement of central heater, steel canister, bentonite blocks/bricks and multiple sensor arrangement.

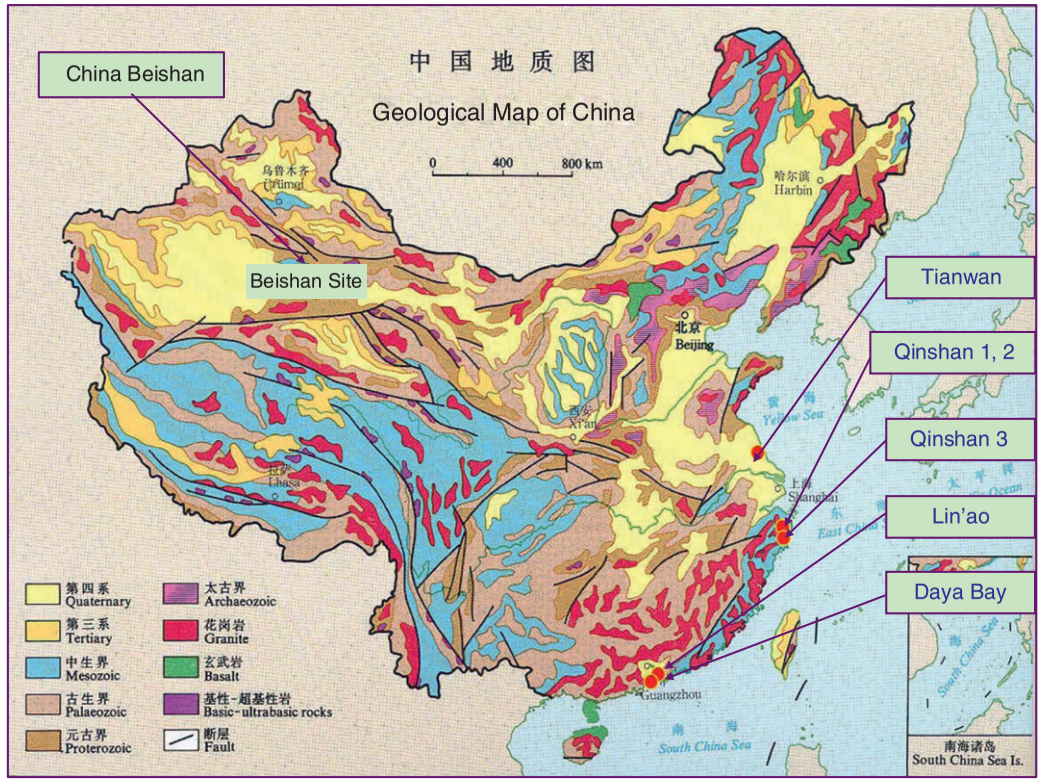

Plate VII (Chapter 22) Beishan HLW geological repository site is in the western part of China, far from any operating NPP.

23

Japan: experience of radioactive waste (RAW) management and contaminated site clean-up

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Abstract: This chapter summarizes the current strategy and policy for radioactive waste management in Japan which has been hindered by a lack of public acceptance and of a final high level waste end-point (geological repository). Ongoing decommissioning of several nuclear facilities, including the Tokai-1 NPP, the Advanced Thermal Reactor 'Fugen' and the Plutonium Fuel Fabrication Facility (PFFF), are described.

Key words: radioactive waste treatment, radioactive waste disposal, decommissioning and dismantling, nuclear facilities, policy and strategy, Japan.

23.1 Introduction

This chapter was written largely before the Fukushima accident, details of which and the clean-up programme are included in the next chapter. However, not only nuclear policy but also nuclear safety regulation in Japan is likely to change in the future.

23.1.1 Nuclear energy in Japan

Japan has carried out nuclear power generation research since the middle of the 1950s. A test power reactor, the Japan Power Demonstration Reactor (JPDR), started operation in 1963 and Tokai-1 Nuclear Power Plant (NPP), the first commercial reactor, went into operation in 1966 with a generation capacity of 166 MWe. Currently, about 50 commercial nuclear reactors, predominantly boiling water reactors (BWRs), and pressurised water reactors (PWRs), are in operation, with a total generation capacity of 48,847 MWe. Prior to the Fukushima disaster, about 30% of Japan's electricity came from nuclear power (Plate VIII between pages 448 and 449). Japan will continue to develop nuclear power as a mainstay of non-fossil energy, while placing the highest priority on safety[1].

The Framework for Nuclear Energy Policy (FNEP), which was established by the Japan Atomic Energy Commission (JAEC) as the basics for political measures regarding the use of nuclear power generation and radiation to be promoted by governmental agencies for the next 10 years, was approved by the Cabinet in October 2005[2].

Prior to the events at Fukushima, nuclear energy was expected to continue to contribute to the pursuit of an optimum energy supply mix for Japan. The FNEP specified that nuclear power's share of Japan's total power generation should be maintained at 30–40% or more beyond 2030 and that the nuclear fuel cycle should be promoted[3]. Nuclear power generation is the key base-load power source. After Fukushima, in July 2011, the Energy & Environment Council (Enecan or EEC) was set up by the Cabinet Office to recommend on Japan's energy future to 2050. It is chaired by the Minister for National Policy and will focus on future dependence on nuclear power. In September 2011, Japan's prime minister said he expected the country to reduce its dependency on nuclear power in the medium and long term, and that the government would address the question of those new plants now under construction. He said that the national Basic Energy Policy would be revised from scratch, and that a new strategy and plan to 2030 would be created. He also stated that Japan's ministerial-level Energy and Environment Council would 'thoroughly review nuclear policy and seek a new form'. The review may recommend that nuclear power's contribution to electricity be targeted at 0%, 15%, or 20–25% for the medium term – a 36% option was dropped.

23.1.2 Radioactive waste (RAW) management policy

Radioactive waste (RAW) is generated by the research, development and utilization of nuclear energy at NPPs, nuclear fuel cycle facilities, test and research reactors, universities, institutes, and medical facilities, using accelerators, radioactive isotopes (RI) and nuclear fuel materials. It is essential that activities associated with research, development and utilization of nuclear energy also process and dispose of the RAW in such a way as to prevent any significant effects on the human environment now and in the future.

The generation that has enjoyed the convenience and benefits of nuclear energy assumes the responsibility to expend all efforts for safe disposal of RAW for the next generation. There are four principles for the treatment and disposal of RAW:

1. The liability of generators,
2. Minimization of radioactive waste,
3. Rational treatment and disposal,
4. Implementation based on mutual understanding with the people.

Under these principles, it is important to appropriately classify the wastes and treat and dispose of each classification safely based on the recognition that the wastes may include materials with characteristics that take an extraordinarily long time for the radioactivity to drop to insignificant levels[2].

A near-surface disposal facility already operates for most of the low-level radioactive waste (LLW) generated at NPPs and is operated in Rokkasyo, Aomori-Ken by Japan Nuclear Fuel Limited (JNFL), as a private business, excluding part of the LLW. With regard to near-surface disposal of RI and research wastes, the Japan Atomic Energy Agency (JAEA) will conduct and promote disposal activity in cooperation with the government and other waste generators. As for the remaining LLW, JNFL plans to construct an intermediate depth disposal facility for NPPs and the Nuclear Waste Management Organization of Japan (NUMO) will geologically dispose of transuranic (TRU) wastes. Funds from the owner of the reprocessing plant and mixed oxide (MOX) fuel fabricator, etc., have been accumulating via a levy to pay for geological disposal of TRU wastes since 2009. However, the implementing body for subsurface disposal of LLW, RI and research wastes has yet to be decided.

High-level radioactive waste (HLW), generated during reprocessing spent fuel (SF), is being vitrified and packaged prior to disposal in a geological repository. Research and development for that purpose had been conducted mainly by what was the Power Reactor and Nuclear Fuel Development Corporation (PNC), which was restructured as the JAEA in October 2005 through the Japan Nuclear Cycle Development Institute. The government worked to develop a disposal system taking into consideration these policy guidelines and scientific evidence, and enacted the Specified Radioactive Waste Final Disposal Act in June 2000. NUMO was created in October 2002 as an implementing body for disposal, as specified in the Act. In December 2002, NUMO started 'open solicitation', which encouraged municipalities to consider investigating the suitability of their local area for developing a deep repository for HLW. Meanwhile, electric utilities and others have been accumulating funds for the disposal of HLW.

23.2 Radioactive waste (RAW) management strategy

23.2.1 Sources, types and classification of radioactive waste

In Japan, RAW is categorized as shown in Table 23.1[4]. In May 2007, the Nuclear Safety Commission of Japan (NSC) issued a document which provides for upper bounds of concentration of radioactive elements in waste packages from power reactors and in TRU waste packages. The upper bounds of concentration of radioactive elements are so decided, that the

Table 23.1 Classification of radioactive wastes in Japan

Classification			Origin of	Disposal method
High-level radioactive waste			Reprocessing plant	Deep geological disposal (>300 m)
Low-level radioactive waste	Waste power reactor	Relatively higher level	Nuclear power plant	Sub-surface disposal (50–100 m)
		Lower level		Near-surface disposal with artificial barrier
		Very low level		Near-surface disposal without artificial barrier
	TRU		Reprocessing plant, MOX fuel fabrication plant	Deep geological, sub-surface and near-surface disposal
	Uranium waste		Enrichment plant, fuel fabrication plant and conversion and refining plant	Not yet decided
	RI and research waste		Research facility RI utilizing facility	Deep geological, sub-surface and near-surface disposal

public exposure due to waste packages is well within the reference value, and that the upper bounds conform to the latest knowledge in the international community. Based on these concepts, disposal of RAW is categorized into Category 1 Waste disposal (geological disposal) and Category 2 Waste disposal (sub-surface disposal, near-surface disposal with artificial barrier and near-surface disposal without artificial barrier).

Concerning the waste that does not need to be dealt with as RAW, the NSC has studied the clearance level of radionuclide concentrations and its calculation method, by reference to the ICRP document (Pub. 46, 1985) and IAEA-TECDOC-855, respectively.

23.2.2 Radioactive waste treatment

In Japan, HLW and LLW are generated through nuclear power generation, nuclear industries (enrichment, fuel fabrication, reprocessing, etc.), utilization of RI and research. HLW is only generated from reprocessing plants and consists of liquid waste, which is mainly stripped liquid effluent from the extraction process, and solid waste, which is vitrified products of the liquid waste.[5]

LLW is generated from all nuclear facilities and consists of gaseous, liquid and solid wastes. Wastes from NPPs include gaseous waste (e.g., off-gas from the reactor system and off-gas from the ventilation system of the reactor building), liquid waste (e.g., effluents from the reactor cooling system, floor drains of the reactor building and detergent waste from laundry and hand-wash), and solid wastes (e.g., spent ion-exchange resin, paper, cloth, plastic sheets, tools, pipes, exchanged parts of equipment and filters).

Wastes from nuclear fuel cycle facilities include gaseous waste (e.g., off-gas from each process, vessels, hot cells, glove boxes and building ventilation systems), liquid wastes (e.g., effluents from the off-gas scrubber, the acid recovery process and the solvent washing process, drain from analytical laboratory, solvent waste, floor drain and detergent waste), and solid wastes (e.g., hulls, gloves, vinyl bags, paper, cloth, plastic sheets, spent sampling jugs, tools, exchanged parts of equipment and filters).

Wastes from enrichment and uranium fuel fabrication facilities include gaseous waste (e.g., off-gas from the building ventilation system), liquid waste (e.g., the floor drain and detergent waste), and solid waste (e.g., paper, cloth, tools pipes, exchanged parts of equipments and filters).

Wastes from research facilities, and facilities in which RI are used for medical and industrial purposes, include gaseous wastes (e.g., off-gas from building ventilation systems), liquid wastes (e.g., chemical waste, spent liquid scintillate, the floor drain and detergent waste), and solid wastes (e.g., experimental instruments, syringes, paper, gloves, plastic sheets, tools and

filters). The management of these types of waste in Japan is described in Section 23.2.4.

23.2.3 Storage and disposal of radioactive waste

A large portion of Japan's radioactive waste (about 50%) is stored in the radioactive waste management facilities at the nuclear facilities. About 1,690 canisters of vitrified product and about 380 m^3 of liquid waste as HLW are stored in the reprocessing facilities at Tokai and Rokkasho (interim storage facility of the glass canisters), as of the end of March 2010. About 267,000 m^3 of LLW (excluding used steam generators, spent control rods, disused channel boxes) are stored in all nuclear facilities in Japan as of the end of March 2008. Storage volume of LLW is made up of approximately 144,000 m^3 NPP waste, approximately 25,000 m^3 TRU waste, approximately 9,000 m^3 uranium waste, approximately 65,000 m^3 research waste, and approximately 24,000 m^3 RI waste[6].

The JNFL near-surface disposal facility with engineered barrier systems in place at Rokkasho, Aomori-Ken is in operation for LLW from commercial NPPs and about 219,000 200 L drums have been disposed of as of the end of March 2010. About 1,670 tons of very low level wastes resulting from dismantling of the Japan Power Demonstration Reactor (JPDR) were disposed of at the near-surface disposal facility without engineered barriers at Tokai. This disposal facility has been on hold since October 1997.

23.2.4 Waste treatment practices[7–10]

NPPs

Off-gas waste from NPPs contains mainly short-lived noble gas nuclides. Off-gas treatment is aimed at decay of short-lived nuclides and the removal of aerosol radionuclides. The off-gas treatment system consists of a hydrogen recombiner unit, an activated charcoal retention unit and a filtration unit. The off-gas from the ventilation system is passed through the charcoal filter and the high efficiency particulates air (HEPA) filter to eliminate iodine and aerosol, respectively. The treated off-gas is discharged through a stack after verification that it is under the regulatory limit.

Treatment of the liquid waste from NPPs aims where possible at recycling in the plant system, removing the radioactivity in controlled liquid discharges and eliminating process effluents. The liquid waste treatment system in a BWR is composed of the low conductivity subsystem, the high conductivity subsystem, the detergent waste subsystem, and the solidification subsystem. The liquid waste from processes in BWR is divided into the low conductivity effluent, which has relatively high purification, and the high conductivity effluent, which has relatively low purification. The low

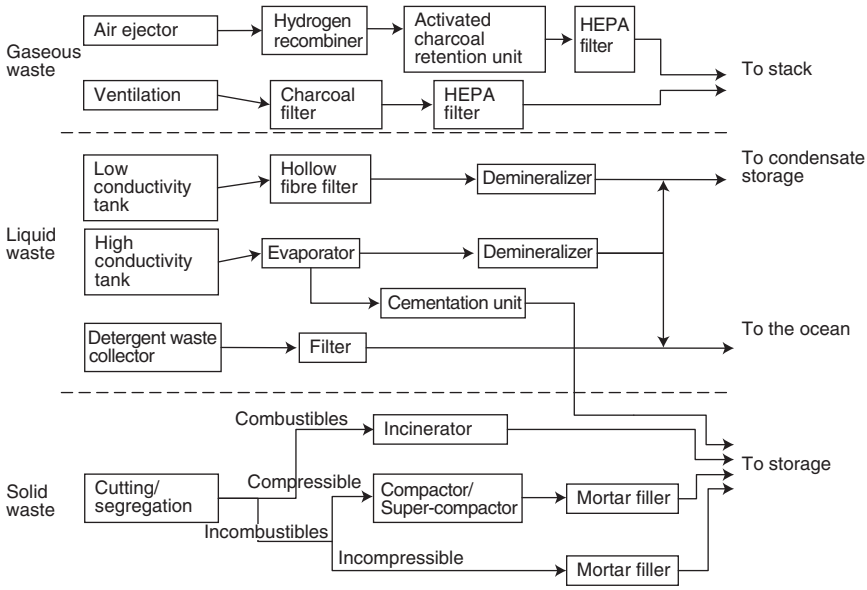
conductivity subsystem collects and processes liquid wastes typically from the equipment drains in the primary cooling system. This waste is filtered through a hollow fibre membrane for removal of insoluble material and demineralized by mixed ion exchange resin for removal of soluble chemicals, and then returned to condensate storage prior to further use as reactor coolant.

The high conductivity subsystem collects and processes liquid wastes from floor drains and effluents from regeneration of the resins. These wastes are concentrated by evaporation, and fed to the solidification subsystem. The distillate is demineralized on the mixed ion-exchange resin, and then returned to condensate storage or discharged to the ocean after verification under regulatory limit. The detergent subsystem collects and processes detergent waste from personnel hand-wash and laundry operations. These wastes are filtered, and then discharged to the ocean.

The solidification subsystem collects concentrated waste in a dedicated tank. This waste is mixed with cement or bitumen, and solidified in 200 L drums. The latest solidification subsystem adopts the dry-pelletizing method in which the concentrated waste is dried with a film evaporator, and dried powder mixed with binder is pelletized by a granulator. This method gives a high waste reduction volume compared to cementation/bituminization.

The liquid waste treatment system for PWR is similar to that for BWRs. PWR employs the recoverable effluent subsystem corresponding to the low conductivity subsystem of BWR. The recoverable effluent containing boric acid from the primary coolant system is demineralized, and then treated by evaporation to separate water and boric acid solutions for further use. Other subsystems are similar to those in BWRs.

Treatment of the solid waste from NPPs is aimed at stabilization and volume reduction for storage and conditioning prior to disposal. The solid waste treatment system is constructed typically of the pre-treatment subsystem, the incineration subsystem for combustible material, the compaction subsystem for incombustibles and the conditioning subsystem. The pre-treatment subsystem is composed usually of cutting and segregation. Large wastes are cut into small pieces appropriate for compaction/packing. Wastes are sorted into combustible, incombustible, compressible and incompressible wastes. Combustible wastes are burned typically in an excess air-type incinerator, and the incinerated ash is placed in a 200 L drum for storage. The compaction subsystem usually makes waste compacts of compressible and incombustible wastes with a compressing force between 50 kN and 3 MN. For higher reduction ratios, the super-compactor or the melting system is adopted in some NPPs. Waste compacts and incompressible wastes are placed in 200 L drums, and then filled with mortar in the conditioning subsystem for disposal. Figure 23.1 shows a typical treatment flow for BWR wastes.



23.1 Typical waste treatment flow for BWR waste.

Nuclear fuel cycle facilities[11,12]

Treatment of the gaseous waste from nuclear fuel cycle facilities removes aerial radioactive particles and gaseous radioactive nuclides before discharge of the gaseous effluent into the environment. In reprocessing plants, the gaseous waste is filtered through scrubbers, Ag-zeolite/Ag-silica-gel filters for iodine and HEPA filters, and then discharged through a stack after radioactivity measurement to ensure it is below regulatory limits. In MOX fuel fabrication plants, only aerial radioactive particles are generated. The off-gas is passed through HEPA filters, and discharged through a stack after measuring radioactivity.

Treatment of the liquid waste from reprocessing plants removes radioactivity in controlled liquid discharges and eliminates process effluents. The liquid waste treatment system is composed typically of the high active liquid subsystem, the intermediate active liquid subsystem, the low active liquid subsystem, the solidification subsystem and the solvent waste subsystem. The high active liquid subsystem collects and processes typically raffinate from the separation/extraction process. This waste is concentrated by evaporation, vitrified and stored in a dedicated interim facility.

The intermediate active liquid subsystem collects and processes effluents from the acid recovery process, the solvent washing process, the off-gas scrubber, etc. This waste is concentrated by evaporation, and then the distillate is fed to the low active liquid subsystem.

The low active liquid subsystem collects and processes floor drain liquids, the detergent waste, etc. These wastes are distilled, then filtered and discharged into the ocean after activity measurement. The concentrated waste is fed to the solidification subsystem.

The solidification subsystem collects and processes the concentrated wastes from the intermediate active liquid subsystem and the low active liquid subsystem. These wastes are adjusted to appropriate pH and concentrated by flocculation/ultrafiltration, and then solidified with cement in the Tokai reprocessing plant of the JAEA Tokai Research and Development Center. In the spent fuel reprocessing plant in Rokkasho village (Rokkasho reprocessing plant), these wastes are dried with a film evaporator and pelletized with a granulator. The processed solid wastes are stored in the facilities.

The solvent waste subsystem collects and processes waste solvent from the solvent washing process. This waste is solidified with epoxy resin at Tokai and hydrothermally solidified after pyrolysis at Rokkasho.

Treatment of the solid waste is implemented with the aim of volume reduction for storage, because a disposal facility is not yet available in Japan for TRU waste. The combustible wastes are incinerated, and the incinerated ash is placed in 200 L drums for storage. The non-combustible wastes are placed directly in appropriate containers, and stored at the facilities. In Tokai MOX fuel fabrication plant, plastics and polyvinyl chloride (PVC) are also incinerated in a dedicated incinerator.

Enrichment plants

Treatment of the gaseous waste from enrichment plants removes fluoride and radioactive particles before discharge into the environment. The off-gas from centrifuges is typically filtered with NaF filters, alumina filters, and HEPA filters, and then discharged through a stack after radioactivity measurement.

In the enrichment plants, small amounts of liquid waste are generated from floor drainage and detergent wastes. These wastes are typically treated by flocculation using a flocculate agent such as polyaluminum chloride, and discharged into the environment.

Treatment of the solid waste is aimed at volume reduction for storage. Combustible wastes are incinerated, and then placed in 200 L storage drums. Incombustible wastes are placed directly in appropriate containers, and stored at the facilities.

Research facilities[13]

Treatment of the gaseous waste from research facilities is performed to remove radioactive particles before discharge into the environment. The

off-gas from the ventilation system is passed through a HEPA filter, and then discharged through a stack after radioactivity measurement.

Small amounts of liquid waste are generated from chemical drains, floor drains and detergent waste. These wastes are treated to remove radionuclides by flocculation or evaporation, and discharged into the environment. Pre-treatment such as neutralization is performed before treatment as needed.

In many small laboratories, solid wastes are placed in containers for storage without treatment. However, large institutes, in which many solid wastes are generated, treat the solid waste to reduce its volume for storage. The solid waste treatment system consists typically of pre-treatment, incineration and super-compaction. The pre-treatment is composed of cutting and segregation. Large wastes are cut into small pieces appropriate for compaction/packing. Solid wastes are sorted into combustible wastes and non-combustible wastes. The non-combustible wastes are further sorted into compressible and incompressible wastes. The combustible wastes are burned in excess air-type incinerators, and then stored. The compressive wastes are compressed with 20 MN force at the super-compactor. The waste compacts and incompressible wastes are placed in 200 L storage drums.

Medical, industrial and research laboratories using RI (RI waste)

Radioactive liquid and solid wastes from utilization of RI are exclusively collected and treated by the Japan Radioisotope Association. Organic liquid waste and flammable solid wastes are treated by incineration, and stored in suitable containers. Inflammable solid wastes are compressed with a compactor, and placed in 200 L drums.

Technological development

Development of treatment technology for liquid wastes is mainly aimed at solidification of the concentrated liquid waste to reduce the product volume ratio. Cementation technology was originally adopted while bituminization, plastic solidification with unsaturated polyester resin, and improved cementation technologies were developed later. The latest solidification treatment adopts the dry-pelletizing method combining a film evaporator and a granulator. The resulting granules are mixed with Portland cement and solidified in 200 L drums. This method gives a high waste volume reduction compared to the early cementation method.

Technology development of solid waste treatment has largely focused on incineration technology and volume reduction technologies for miscellaneous waste. Incineration for chlorine-containing materials including PVC using an incinerator with a water-cooled cylindrical chamber has been

developed. This incinerator has an off-gas system as a countermeasure against dioxin. An incinerator with an ash melting system has also been developed with a high temperature chamber to burn and melt simultaneously or an incinerator and a separate melting furnace. This type of incineration system can treat flammable and non-flammable materials producing slug granules with high volume reduction ratio.

For non-combustible wastes, super-compaction technology and melting technology have been developed. A super-compaction system with over 10 MN compressive force has been developed using a triaxial compressive or a uniaxial compressive machine with a drawing mould for direct encapsulation to 200 L storage drums. Both high-frequency induction or plasma heating furnaces are used in NPP for melting. The high-frequency induction furnaces use a disposable crucible, which can be placed directly in a 200 L drum. Some treatment technologies such as vitrification, steam reforming, etc., for iodine filters and ion-exchange resins are under development.

23.2.5 Radioactive waste disposal[14]

High-level radioactive waste

In line with the Specified Radioactive Waste Final Disposal Act, final disposal facilities are planned for the geological disposal of HLW and are scheduled to start operation in the 2030s through the following three-step selection process: selection of preliminary investigation areas, selection of detailed investigation areas, and selection of the final disposal facility areas. When local governments wish to volunteer for 'areas to be investigated as to the feasibility of constructing final repository of HLW', it is important that the implementor (NUMO), the government and the utility companies give sufficient understanding and awareness to the local residents about the advantages and disadvantages of the final repository and various sectors of the local community, including local government. The government, research and development (R&D) institutions and NUMO, while giving due consideration to their own roles and in close partnership, are expected to consistently promote R&D into HLW geological disposal. NUMO is expected to safely implement the final HLW disposal project and systematically perform technical development to improve the economics and efficiency of the disposal activities. R&D institutions, led by the JAEA, through utilization of underground research facilities, continue to conduct research on underground geology, basic R&D towards improved reliability of geological disposal technology and safety assessment methods, and for safety regulations.

While being aware of overseas knowledge and experience, it is important to develop and maintain an advanced knowledge base that supports final

repository projects and safety regulations, as well as to appropriately reflect it in NUMO's final disposal projects. To this end, the government and R&D institutions work together to survey the entire Japanese waste management programme systematically and efficiently. R&D institutions such as JAEA, Radioactive Waste Management Funding and Research Center, etc., need to cooperate with the government and NUMO in activities to improve the understanding and awareness of society at large. Furthermore, it is necessary for the government to develop specific rules concerning safety regulations based on the progress of these R&D activities.

Geological disposal of radioactive wastes containing transuranium elements

Some LLW containing TRU elements needs to be disposed of geologically. If some TRU waste targeted for geological disposal can be buried together with HLW (co-disposed), the number of repository sites may be reduced, improving economic efficiency. Based on assessment of the influence of TRU and HLW co-disposal on the integrity of the disposal site, the government should then consider necessary measures, including the nature of an implementing body and its own involvement.

LLW from overseas reprocessing consigned by Japan will gradually be returned from France and the UK. French reprocessing firms suggest changing the solidification method from embedding in bitumen to vitrification, while UK reprocessing companies will embed the LLW in cement for geological or disposal with institutional control. In the latter case, the waste returned to Japan is HLW (vitrified waste) with equivalent levels of radioactivity to the LLW exported. In light of these suggestions, it is expected that the number of shipments can be reduced and storage facilities in Japan for LLW awaiting final disposal can be downsized. Thus, the government, in response to discussions with the operators, will assess the benefits of waste treatment by the new solidification methods, suggested by France, and of the conversion indexes of waste, as suggested by the UK. If these suggestions are found to be acceptable, the government should discuss the institutional issues.

Radioactive wastes for disposal with institutional control

Methods of disposal with institutional control include near-surface disposal without artificial barriers, near-surface disposal with artificial barriers and sub-surface disposal with artificial barriers. Near-surface disposal with artificial barriers is already used for LLW generated in commercial nuclear reactor facilities. Near-surface disposal without artificial barriers is being partly implemented while the reactor operators improve safety regulations

on the remainder of the wastes. Operating entities are conducting studies and tests on sub-surface disposal with artificial barriers. Based on the results, it will be necessary to discuss the establishment of the framework, including safety regulations.

The current status of management of other LLW is described in Section 23.1.2.

23.3 Spent fuel management strategy, practice and issues[15]

In accordance with the basic principle in the Framework for Nuclear Energy Policy, the 'Act for Deposit and Administration of Reserve Funds for Reprocessing of Spent Fuel from Nuclear Power Generation' was established requiring operators to place funds for spent fuel reprocessing in a fund administration corporation. The objective of 'the Act' is to ensure the proper implementation of spent fuel reprocessing, disposal of radioactive wastes generated from reprocessing and decommissioning of the reprocessing facilities. The reserve fund held by the 10 utility companies at the end of March 2007 was a 1,390 billion yen. As a part of its waste management plans, the Ministry of Economy, Trade and Industry (METI) designated the 'Radioactive Waste Management Funding and Research Center' as a non-profit 'fund administration corporation' (October, 2005) that is supervised by the METI through supervisory orders and on-the-spot inspection.

SF generated in power reactors is sent to reprocessing facilities after a period of on-site cooling and storage. SF has historically been reprocessed overseas in accordance with contracts with British and French companies, with the exception of a portion reprocessed by the Tokai reprocessing plant of the JAEA. However, considering the national need, JNFL constructed the Rokkasho reprocessing plant, based on operational experience accumulated at the Tokai reprocessing plant and on overseas technologies and experience. The plant underwent active testing using SF in 2008 and started operation in 2008. Storage of SF in the plant storage facility started in 1999, and export of SF to foreign reprocessing plants ended in July 2001.

The Law for Regulation of Nuclear Source Material, Nuclear Fuel Material and Nuclear Reactors (Reactor Regulation Law) was amended in 1999 to incorporate provisions on interim SF storage. Tokyo Electric Power Company and Japan Atomic Power Company (JAPC) jointly established the 'Recycle Fuel Storage Company' to prepare for commercial operation of the first interim fuel storage facilities planned for 2010. In March 2007, the company applied for a licence to construct and operate the Recycle Fuel Storage Center at Mutsu city, Aomori Prefecture, and the licence application is now undergoing the safety examination.

SF from research reactor facilities has been, and is to be, returned to the US, UK or France, or is to be reprocessed or stored in Japan.

23.4 Decommissioning strategy, practice and issues[16–20]

23.4.1 Decommissioning strategy

The basic policy for decommissioning commercial NPPs was established by the JAEC in 1982. It states that retired commercial NPPs should be dismantled as early as possible after shutdown and the site should be effectively re-used for next generation NPP. The Framework for Nuclear Energy Policy issued by the JAEC states that it is the operator's own responsibility, but under government regulation, to carry out decommissioning of a nuclear facility, ensuring safety, while obtaining local communities' understanding and cooperation.

The regulatory policy for dismantling or decommissioning reactor facilities has been discussed by the NSC. To ensure safety during decommissioning of commercial NPPs, the regulation was implemented by applying existing provisions in the Reactor Regulation Law by the operators. To date, decommissioning of reactor facilities has been implemented at facilities such as the JPDR of the JAEA and the Tokai-1 NPP of JAPC, the development and application of dismantling technologies have progressed, and know-how for decommissioning has been accumulated. The NSC examined the idea of ideal safety regulation, based on the experience of decommissioning of nuclear facilities. It also took into consideration the features of nuclear facilities post-termination and the level of potential risks.

The Decommissioning Safety Subcommittee has investigated the appropriate regulation systems for decommissioning, based on regulatory experiences of decommissioning reactor facilities. The Decommissioning Safety Subcommittee proposed the decommissioning regulations as:

- replacing dismantling notification by licensee, to approval of the licensee's decommissioning plan by the regulatory body,
- implementation of decommissioning as approved in the decommissioning plan,
- completion of decommissioning is confirmed by the regulatory body and after confirmation of the completion of decommissioning, the operating licence becomes ineffective,
- the regulatory activities during the decommissioning process should be changed in accordance with the changes of functions of facilities and safety operation activities as the decommissioning proceeds.

On the basis of such recognition, the Reactor Regulation Law was amended in 2005. A licensee applying for approval of decommissioning has to submit a decommissioning plan that describes, for example dismantling methods, radiation controls, safety assessment and the financial plans. The regulatory body approves the decommissioning plan after examining its conformity with technical standards. At the final stage of decommissioning, the licensee submits a document that describes the implementation status of dismantling, management of contaminated materials and the final distribution of contamination and requests the regulatory body's confirmation. The decommissioning is completed after the regulatory body confirms that the measures for radiation hazard prevention are no longer necessary and management of contaminated materials is completed.

23.4.2 Decommissioning practice and issues

Nuclear facilities in the process of being decommissioned in Japan include Tokai-1 NPP of the JAPC, the Advanced Thermal Reactor 'Fugen' of the JAEA and the Plutonium Fuel Fabrication Facility (PFFF) of the JAEA. Hamaoka Nuclear Power Station Reactor's No. 1 and 2 of Chubu Electric Power Company shut down in January 2009 and their decommissioning plans were approved by the METI in November 2009.

Tokai-1

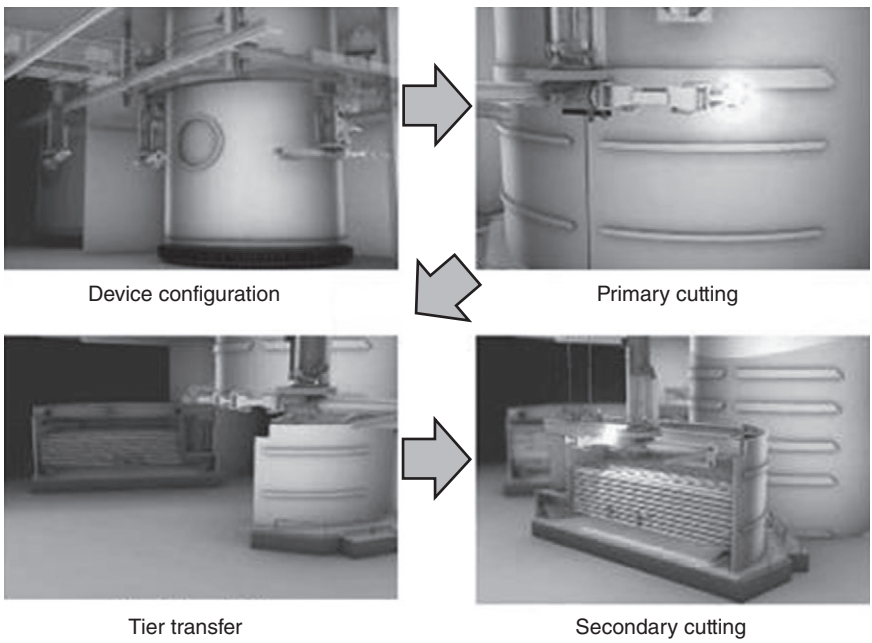
Tokai-1 NPP (GCR) is a graphite-moderated, gas-cooled reactor. Tokai-1 NPP started commercial operation in 1966. However, it has disadvantages from an economic standpoint because the carbon dioxide GCR has a relatively low power output for the volume of the reactor. This raises the cost of electricity generation compared with light water reactors. Tokai-1 NPP was finally shut down in 1998 after it was defuelled and all fuel elements were shipped off-site for reprocessing by 2001. The reactor area was stored in a safe condition for 10 years after final shutdown to reduce radioactivity. During safe storage, conventional facilities outside the reactor area are removed to secure a transportation route for dismantling wastes, and also to create space for new waste conditioning facilities. Starting with peripheral equipment outside the reactor area, Tokai-1 NPP is being demolished in stages. Equipment in the reactor area will be dismantled and removed after being securely stored until radioactivity has decayed to an allowable level. Finally, the site would be able to be re-used for a new NPP. Dismantling activities initiated in 2001 and during the first five years, conventional facilities, such as the turbine system were removed. Cartridge cooling pond (CCP) water was also drained and the CCP was cleaned up for clearance activities.

Four steam raising units (SRUs) have been removed since 2006. The SRUs are 24.7 m in height, 6.3 m in diameter and within the SRUs are radioactively contaminated and complicated structures. Jack-down methods and a remote dismantling system were developed for workers' safety and to minimize the extent of the contaminated areas. The SRUs are removed with the system remotely in turn from bottom while lifting them with large jacks. The system enables cutting and holding not only of the SRU body but also other internal parts of the SRUs. Figure 23.2 shows images of cutting work on the SRU. The jack-down method prevents the need to work in high places and restricts the radiation controlled area to the bottom area of the SRU.

Fugen

Fugen NPP (ATR, 165 MWe) is a heavy water-moderated, boiling light water-cooled, pressure tube-type reactor. Fugen NPP began operating in March 1979, finally shut down in March 2003, and its decommissioning plan was approved in February 2008. Fugen NPP dismantling was separated into the following four periods.

1. Spent fuel transfer period. SF will be transported to Tokai reprocessing plant, and heavy water will be transported to Canada for re-use at



23.2 Cutting working images of the SRU.

CANDU reactors. Less contaminated equipment such as turbines will be dismantled, while some related systems for SF storage remain operational.
2. Peripheral facilities dismantling period. After SF transportation is complete, the related SF storage systems are dismantled and the peripheral reactor equipment will be dismantled to enable installation of remotely operated dismantling machines.
3. Reactor core dismantling period. The reactor core by dismantled by remote operation underwater, and it is expected that the exposure dose in the dismantling activities will be minimized to the equivalent dose of an annual inspection during the plant operation. In this period, both dismantling of all contaminated equipment and decontamination of buildings will be carried out to release the radiation-controlled area.
4. Building demolition period. In this period, both released buildings and non-contaminated buildings will be demolished by conventional methods.

After the approval of the programme, decommissioning was initiated. SF has been transferred to the Tokai reprocessing plant, and heavy water has been transported to Canada. Dismantling of the turbine facility was started in parallel. Two of five feedwater heaters and main steam lines were dismantled. Experience of cutting technologies and relevant data such as total manpower have been accumulated for future work.

The reactor core of Fugen NPP has a complicated configuration arising from its pressure tube-type structure. The pressure tube and the calandria tube are made of zirconium alloy which can be combustible in powder form. Also, they have been highly activated during operation. It is thus planned to dismantle the core structure underwater for shielding radiation, to prevent airborne dust and for fireproof cutting.

Uranium refining and conversion plant (URCP)

The uranium refining and conversion plant (URCP) at Ningyo-toge was constructed in 1981 to demonstrate refining and conversion of yellow cake (or uranium trioxide) to uranium hexafluoride via uranium tetrafluoride. There are two different types of refining processes in the URCP. One is the wet process for converting natural uranium and the other is the dry process for reprocessed uranium.

Dismantling of the dry process facilities began in March 2008. The basic strategy concerning plant dismantling was to optimize the total labour costs and minimize the radioactive wastes generated. The basic schedule for dismantling is as follows.

- Phase 1: removal of large equipment or processes involving uranium hexafluoride,
- Phase 2: removal of the greater part of the utilities connecting the main process of URCP,
- Phase 3: removal of equipment of main process,
- Phase 4: removal of ventilation systems.

The majority of equipment will be dismantled, except for building decontamination, by 2013. A large amount of fluidization media had been stored in tanks held underground in the URCP. The fluidization media is composed of small aluminum pellets which absorbed uranium oxides or unreacted uranium tetrafluoride used for the fluorination reaction. They therefore contain high levels of uranium and thorium as progenies of U-232. Among its progenies, Tl-208 is a high gamma emitter, so some external exposure will arise in handling the fluidization media.

Uranium enrichment demonstration plant (UEDP)

The uranium enrichment demonstration plant (UEDP) in Ningyo-toge was used to demonstrate uranium enrichment by the gas centrifuge (GCF) method, and was operating continuously from 1988 to 2001. As a result, significant uranium was deposited in the equipment mainly as intermediate uranium fluorides. System chemical decontamination using IF_7 gas was proposed as an efficient decontamination method. The secondary waste characteristic of IF_7 treatment is IF_5 and minor adsorbent. In addition, IF_5 is easy to convert to IF_7 and re-use for system decontamination. The IF_7 treatment technique is performed at room temperature and very low pressures such as 10–45 hPa. Secondary reaction is insignificant in IF_7 treatment except for the reaction between IF_7 gas and the intermediate uranium fluoride. The weights of uranium deposited in the cascades were approximately 700 kgU per cascade before IF_7 treatment. The IF_7 treatment period for each cascade is 60 days applying the near-optimal processing conditions. More than 96% of uranium was recovered from the cascade system. As a result, the U radioactivity of the main parts of the GCF fell to 1.0 Bq/g and below.

Plutonium fuel fabrication facility (PFFF)[21,22]

The plutonium fuel fabrication facility (PFFF) operated from 1972 to 2002 for fabricating MOX fuels for Fugen and the experimental Joyo fast breeder reactor (FBR). The decommissioning and dismantling (D&D) project for the PFFF is divided into the following four phases:

- Phase 1 (up to 2010): stabilization and shipment of nuclear material in the facility. Choose decontamination and volume reduction techniques.
- Phase 2 (2010–2015): D&D planning and adaptability tests.
- Phase 3 (2015–2020): size reduction of equipment and glove box. R&D programme carried out.
- Phase 4 (2020–2035): re-use of buildings for waste storage.

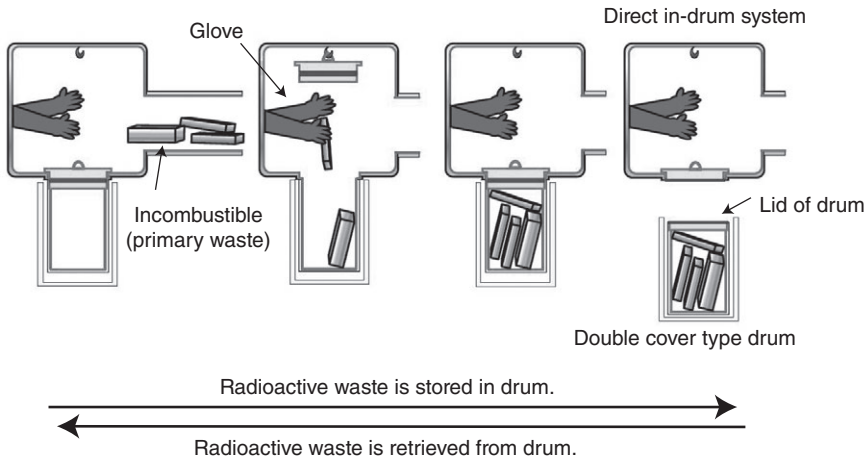
An issue relating to the accumulation of special nuclear material became apparent in the 1990s in this facility. Eight glove boxes in the facility had to be replaced by those with an improved automated fuel fabrication system and residuals recovery system. In order to dismantle these glove boxes, it was necessary to have a more durable containment structure than that of the plastic enclosure, commonly used at the time. To circumvent these issues, the glove box dismantling facility, a centralized decommissioning workshop to dismantle glove boxes, was developed. The purpose of the workshop is to safely dismantle the after-service glove boxes and recover the fuel residuals from the glove boxes. The basic concepts of the workshop are as follows:

1. The workshop has the functionality of a glove box. To prevent the spread of contamination, the level of the internal pressure is kept around 300 Pa in gauge pressure negative to the surrounding room pressure.
2. The workshop is installed in a room in the basement of the plutonium fuel production facility (PFPF) and used for glove box dismantling repeatedly.
3. Remote-controlled devices are installed in the workshop to reduce the radiation dose to which workers are exposed.

The activity undertaken was of both remote and hands-on type size reduction. The data and knowledge will be reflected in the planning of the D&D project for PFFF.

Technological developments to reduce secondary waste generation are being carried out. Dismantled equipment is cut and wrapped in plastic sheets and packing tape, and stored in 200 L drums. The amount of packaging material (secondary waste) sheets may be about 20% of the volume of dismantled materials. In addition, the packaging activities are performed by workers wearing airline suits. These suits are also secondary wastes. In addition, waste treatment facilities will remove the packaging materials from the dismantled equipment which must then be sorted.

To reduce waste treatment work and the amount of secondary waste, a direct in-drum system for RAW management has been developed. The direct in-drum system can be stored directly in a double-skin drum without packaging. In addition, RAW stored in the drums can easily be retrieved



23.3 Direct in-drum system.

from them. To prevent the leakage of radioactivity during storage and retrieval of RAW, the lid of the double-skin drum and of the direct in-drum system are connected by a gasket. The direct in-drum system (Fig. 23.3) is attached to the glove to be used for waste storage.

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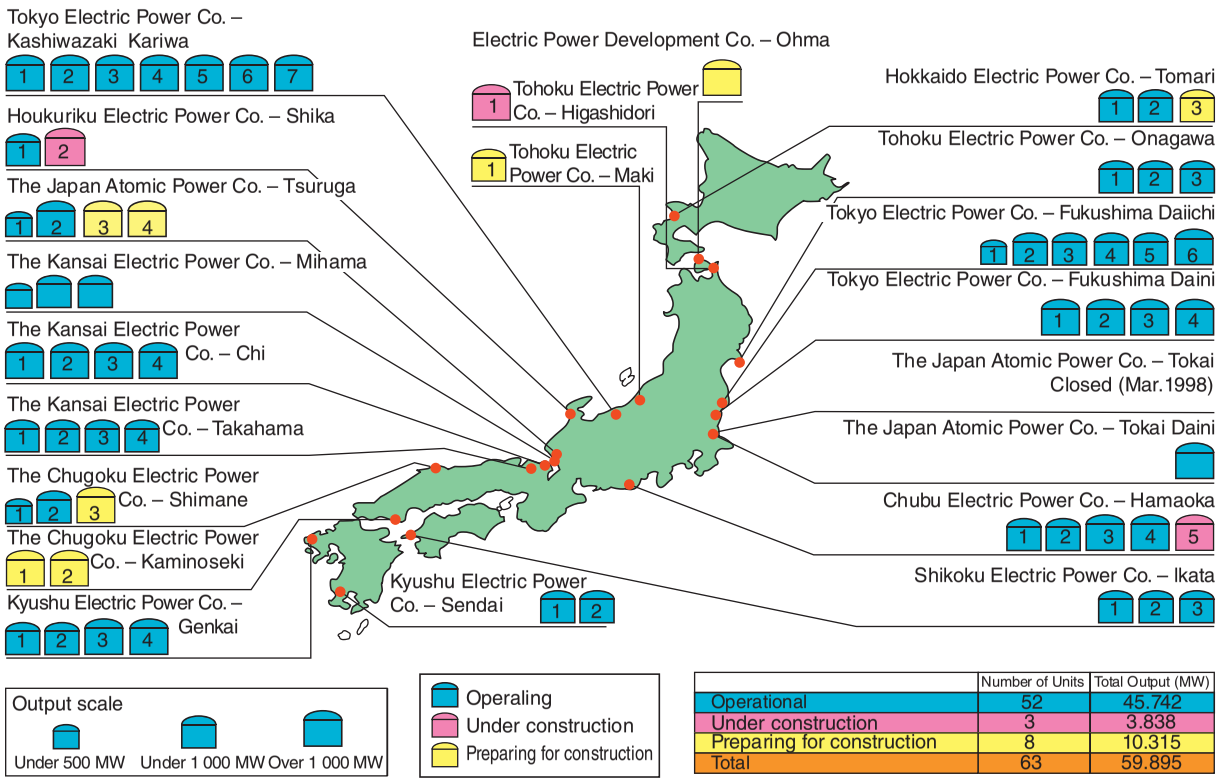

	Number of Units	Total Output (MW)
Operational	52	45.742
Under construction	3	3.838
Preparing for construction	8	10.315
Total	63	59.895

Plate VIII (Chapter 23) Map of Japan showing location of its NPP fleet (prior to shutdowns caused by Fukushima).

24

Fukushima: The current situation and future plans

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Abstract: This chapter describes the Fukushima Daiichi nuclear power plant (NPP) accident starting with the reactors' location and sequence of events which caused the accident. The amount of radioactive materials released and its composition, dispersion of radioactive materials over land and sea, contamination effects on food and the environment and radiation effects on human health are all addressed along with the current clean-up programme and future plans. However, it is expected that it will take 30–40 years to decommission the damaged facilities.

Key words: Fukushima, BWR reactors, contamination, dispersion, clean-up.

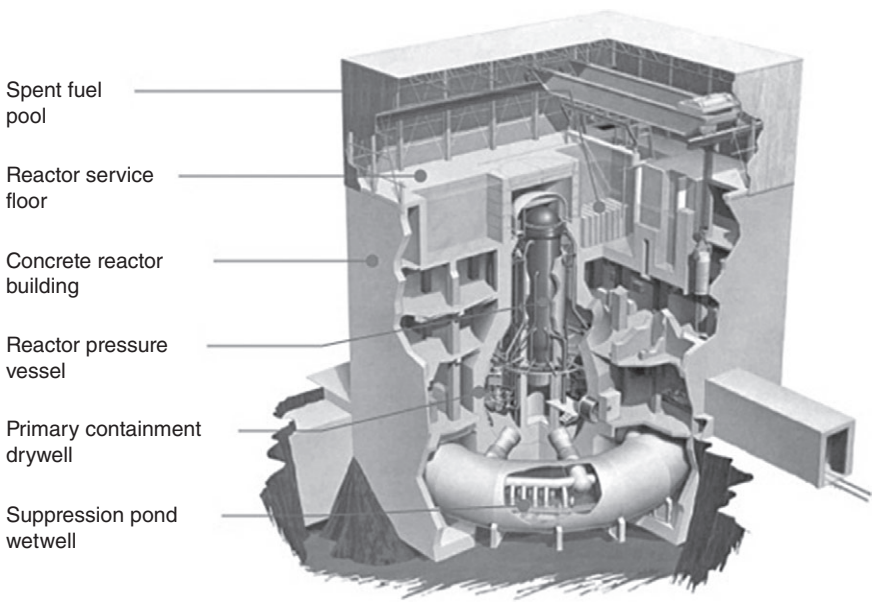
24.1 Introduction

The biggest nuclear disaster since Chernobyl in 1986 occurred at the Fukushima Daiichi nuclear power plants (NPP) on 11 March 2011 (Plate IX between pages 448 and 449). Fukushima was shaken by an earthquake measuring magnitude 9.0 on the Richter scale followed by a massive 14 m tsunami, which caused the deaths of over 20,000 people living along the eastern coast of Japan, and led directly to the shutdown of three reactors in operation. However, the 6 NPP on the Fukushima site were designed on the basis of an earthquake equivalent to magnitude 8.2 and a 5.7 m tsunami. The tsunami caused the unanticipated total loss of power supply because of the loss of the backup generators due to flooding from the tsunami. This power supply is necessary for cooling water circulation for residual heat removal from the reactor cores, and its loss is what eventually lead to severe damage [1] to the cores in the first three days along with degradation arising from the use of seawater for emergency cooling. As a result, high amounts of radioactive material were released into the atmosphere although, unlike at Chernobyl, only causing predominantly regional-scale contamination by radionuclides. The Japanese authorities announced an official 'cold shutdown condition' in mid-December, as the reactor temperatures had fallen

to below 80°C at the end of October 2011 [2]. Apart from cooling, the ongoing task remains to prevent further release of radioactivity, particularly in contaminated water which has leaked from the three units. According to the Japanese government, the total amount of radioactivity released to date is approximately one-tenth that released during the Chernobyl disaster. However, the full extent and level of radioactive contamination remain unclear.

24.1.1 Fukushima Daiichi reactors

The Fukushima Daiichi reactors are six boiling water reactors (BWR) of an early 1960s design supplied by GE, Toshiba, and Hitachi with power ranges from 460 MWe to 1,100 MWe. They came into commercial operation between 1971 and 1975. Four units (1–4) are of Mark-I type containment, which is the first generation of BWR design (Fig. 24.1). At the time of the accident, the units and central storage facility contained the fuel assemblies as shown in Table 24.1.



24.1 Mark-I type BWR (equivalent to units 1–4 of Fukushima Daiichi NPP) [1].

Table 24.1 Numbers and types of fuel assemblies [3]

Location	Unit 1	Unit 2	Unit 3	Unit 4	Unit 5	Unit 6	Central storage
Fuel type	UO_X	UO_X	UO_x/MOX	UO_X	UO_X	UO_X	UO_x/MOX
Core-loaded fuel assemblies	400	548	548	0	548	764	0
New fuel assemblies	100	28	52	204	48	64	N/A
Spent fuel assemblies	292	587	514	1,331	946	876	6,375

MOX = Mixed Oxide fuel. N/A = not available.
Tanabe [4] has estimated that the unit 1 core contained 78.3 tons of uranium dioxide, 32.7 tons of zirconium, 12.5 tons of steel, 590 kilos of boron carbide and 1 ton of inconel.

24.1.2 Tsunami damage to the reactors

The sequences of events in the reactor accident were as follows:

1. Fukushima was shaken by an earthquake measuring magnitude 9.0 on the Richter scale; however, the six NPPs were designed on the basis of an earthquake equivalent to magnitude 8.2.
2. The three units in operation, units 1, 2 and 3, automatically went into SCRAM (sudden shutting down of a nuclear reactor, usually by rapid insertion of control rods), which was triggered by detecting the high earthquake acceleration. Following the total loss of off-site power, emergency power generators automatically started to supply electricity.
3. The standard post-shutdown cooling modes started up to remove the decay heat. This residual heat must be removed to prevent the nuclear fuel, mainly UO_2, cladding metal, and supporting structural elements from melting in the core of the reactor. The melting point of UO_2 is approximately 2,900°C, while those of cladding and supporting parts are in the range of 1,300–1800°C.
4. About 45 minutes after the earthquake, tsunami waves variously hit the units, destroying seawater pumps for the residual heat removal system and many of the emergency generators. Eventually, this lead to the total loss of the electricity that powered the water pumps used to maintain cooling water circulation around the reactor cores. The spent fuel (SF) storage pools suffered the same problem.
5. In spite of the performance of various emergency core cooling systems, as well as trials to vent the reactor vessel enabling water injection from outside, the core eventually became uncovered by cooling water. Along with the increase in temperature of the uncovered fuel, the reaction of

cladding material with steam to generate hydrogen proceeded rapidly, and the fuel started melting leading to core destruction through meltdown.

6. According to the results of the simulation calculation conducted even with insufficient records of the instrumentation, most of the core is believed to have melted in unit 1. In units 2 and 3, much of the fuel apparently melted but to a lesser extent than in unit 1 and dropped to the bottom of the pressure vessel. It is considered that a certain part of the fused fuels and structural materials flowed out from the reactor pressure vessels (RPVs) into the primary containment vessels (PCVs).
7. During the severe accident process, appreciable amounts of volatile radionuclides (typically these are noble gases, cesium and iodine) are considered to have evaporated. They must have escaped from the RPV into the PCVs, and finally escaped via cracks or openings made under the severe conditions.
8. Hydrogen explosions occurred in units 1, 3 and 4, and these seriously damaged their operation floors at the top of the reactor building, and also the upper side walls of unit 4.

24.1.3 Reactor cooling

Seawater was pumped into the reactor cores and the SF storage pools for about two weeks, in an effort to cool the fuels. The total amount of seawater injected before March 25, when it was replaced by the injection of newly delivered pure water, was 2842, 9197, and 4495 kL for units 1, 2 and 3, respectively. It was estimated that as much as 32 tonnes of sea salt may have accumulated in the reactor units. Boric acid was added to the cores to function as a neutron absorber to prevent re-criticality by the collapsed fuels [5]. Circulation of the water using the circulation lines was found to be impossible because there were appreciable leakages in reactor and/or containment vessels, through which injected water continuously flowed out from the reactor building to the basement of the turbine building. The concentration of cesium-137 in the contaminated water exceeded the order of magnitude of 10^6 Bq/cm^3. By the middle of June 2011, a new water treatment system to decontaminate the highly contaminated water flowing from the cores had been established, and the circulation injection cooling using this facility was started in late June. This system (described in more detail in Section 24.5) cleanses the highly radioactive water, recovered from the basement of the building, and injects the decontaminated water back to the core. After removing radionuclides, water is desalinated before re-injection, reducing the salt content in the water to less than several ppm as of July 2012. The addition of seawater to the core, though it was unavoidable during

the emergency, raised an issue of the adverse effects of the salt, in degrading of the vessels and other devices, as well as reacting with the fused fuels to form complicated chemical forms of debris.

24.2 Extent and composition of radioactive material released

Radioactive materials have been released to the environment from the damaged cores of units 1, 2, and 3. Grambow and Poinssot [6] considered the extent of core damage, suggesting from the available evidence that not only the UO_2 in the fuel but also the zircaloy cladding and steel melted forming a quenched melt (also seen at Three Mile Island and Chernobyl) called corium. They believe substantial amounts of volatile fission products (FPs), such as Cs and I, were released during melting, but that the less volatile FPs and actinides were incorporated into the corium. This corium thus contains most of the most radiotoxic species and presents a very large and long-term challenge. The escape of radionuclides from the damaged cores occurred both through atmospheric release and through water leaks to the sea. It is considered that the major part of the atmospheric release was by the unexpected leak of the containment vessel, whilst deliberate venting through the suppression chamber to reduce gaseous pressure gave rather limited result. A controlled release of very low-level contaminated water from the central waste treatment facility (ca. 4 Bq/cm^3) to the sea was done once, but it has little significance in terms of the total amount of contamination. The major release to the sea was by (1) fall-out of the atmospheric release to the sea, (2) carry over by rainwater, and (3) leakage of highly contaminated water via underground cable (sub-drain) pits.

The extent of radionuclide release to the atmosphere has been evaluated several times by government officials and Tokyo Electric Power Company (TEPCO), but it is still very much an estimate [5–13]. This can be seen in Table 24.2 which compares the estimated release of cesium-134, 137 and iodine-131 in March 2011 by several estimation studies. The differences in the data arise from the different assumptions used about the status and progress of the accident, due to unreliability in instrumentation data as well as in the monitoring data under the severe conditions. The estimate by TEPCO, which showed larger values than government authorities, was obtained by comparing the result of dose monitoring as well as of the analysis of radioactivity deposition onto surfaces, and the results of environmental dispersion calculation code. The calculation is based on an assumed rate of evolution of radioactive particles, and thus still has some uncertainty.

Cesium-134, 137 and iodine-131 are the nuclides that mostly decide the radiological impact on the public, among which iodine-131 due to its short

Table 24.2 Estimated release of major radionuclides to the atmosphere

Organizations	Released amount of radioactivity (PBq)				
	Noble gas	I-131	Cs-134	Cs-137	INES index*
TEPCO (May 2012)	ca. 500	ca. 500	ca. 10	ca. 10	ca. 900
JAEA and NSC (April and May 2012)	–	150	–	13	670
JAEA and NSC (August 2011)	–	130	–	11	570
JAEA (March 2012)	–	120	–	9	480
NISA (April 2011)	–	130	–	6.1	370
NISA (June 2011)	–	160	18	15	770
NISA (February 2012)	–	150	–	8.2	480
IRSN	2,000	200	30		–
Chernobyl (for comparison)	6,500	1,800	–	85	5,200

JAEA: Japan Atomic Energy Agency; NISA: Nuclear and Industrial Safety Agency, Japan; NSC: Nuclear Safety Commission of Japan; IRSN: Institut de radioprotection et de sûreté nucléaire.

*INES index: Radiologically equivalent value to iodine-131. The International Nuclear and Radiological Event Scale (INES) was developed in 1990 by the IAEA and the OECD Nuclear Energy Agency (OECD/NEA) with the aim of communicating the safety significance of events at nuclear installations [14] and Section 1.4.6). If there is an atmospheric release from a nuclear facility then a radiological equivalence to iodine-131 is calculated using conversion factors. For example, the actual activity of the isotope released should be multiplied by some factors (given in [14]) and then compared with the values given in the definition of each level. An event resulting in an environmental release corresponding to a quantity of radioactivity radiologically equivalent to more than several tens of thousands of TBq of iodine-131 is rated to the highest level 7 according to the INES scale.

half-life is only significant for a couple of months after its release. The estimated amount of the other released radioactive nuclides can be seen in Table 24.3, which was evaluated in June 2011 and corrected in October 2011 by the Japan Nuclear and Industrial Safety Agency (NISA). In a general sense, the nuclides other than cesium-134, 137 and iodine-131 have less radiological impact because of their lower radioactivity, low specific radiological effect, as well as short half-lives. It should be noted that the amount of strontium-90 believed to have been released is about 1/100 of that of the sum of cesium-134 and 137, and that of plutonium-239 is about 10^{-7} of that.

The amount of radioactivity released to the sea via the sub-drain pit was estimated to be 4.7 PBq in total of cesium-134, 137 and iodine-131 [8], which is much less than the atmospheric release. Due to the leak of the water from the sub-drain pit, the radioactivity concentration in the seawater sampled at the discharging point showed as high as 10^5 Bq/L of cesium-137 at the

Table 24.3 Estimated atmospheric emission (in Becquerel; Bq) of radioactive substances according to the core damage assessment of Fukushima units 1–3 [7]

Nuclide	Half-life	Unit 1	Unit 2	Unit 3	Total
Xe-133	5.2 d	3.4×10^{18}	3.5×10^{18}	4.4×10^{18}	1.1×10^{19}
Cs-134	2.1 y	7.1×10^{14}	1.6×10^{16}	8.2×10^{14}	1.8×10^{16}
Cs-137	30.0 y	5.9×10^{14}	1.4×10^{16}	7.1×10^{14}	1.5×10^{16}
Sr-89	50.5 d	8.2×10^{13}	6.8×10^{14}	1.2×10^{15}	2.0×10^{15}
Sr-90	29.1 y	6.1×10^{12}	4.8×10^{13}	8.5×10^{13}	1.4×10^{14}
Ba-140	12.7 d	1.3×10^{14}	1.1×10^{15}	1.9×10^{15}	3.2×10^{15}
Te-127m	109.0 d	2.5×10^{14}	7.7×10^{14}	6.9×10^{13}	1.1×10^{15}
Te-129m	33.6 d	7.2×10^{14}	2.4×10^{15}	2.1×10^{14}	3.3×10^{15}
Te-131m	30.0 h	2.2×10^{15}	2.3×10^{15}	4.5×10^{14}	5.0×10^{15}
Te-132	78.2 h	2.5×10^{16}	5.7×10^{16}	6.4×10^{15}	8.8×10^{16}
Ru-103	39.3 d	2.5×10^{09}	1.8×10^{09}	3.2×10^{09}	7.5×10^{09}
Ru-106	368.2 d	7.4×10^{08}	5.1×10^{08}	8.9×10^{08}	2.1×10^{09}
Zr-95	64.0 d	4.6×10^{11}	1.6×10^{13}	2.2×10^{11}	1.7×10^{13}
Ce-141	32.5 d	4.6×10^{11}	1.7×10^{13}	2.2×10^{11}	1.8×10^{13}
Ce-144	284.3 d	3.1×10^{11}	1.1×10^{13}	1.4×10^{11}	1.1×10^{13}
Np-239	2.4 d	3.7×10^{12}	7.1×10^{13}	1.4×10^{12}	7.6×10^{13}
Pu-238	87.7 y	5.8×10^{08}	1.8×10^{10}	2.5×10^{08}	1.9×10^{10}
Pu-239	24065 y	8.6×10^{07}	3.1×10^{09}	4.0×10^{07}	3.2×10^{09}
Pu-240	6537 y	8.8×10^{07}	3.0×10^{09}	4.0×10^{07}	3.2×10^{09}
Pu-241	14.4 y	3.5×10^{10}	1.2×10^{12}	1.6×10^{10}	1.2×10^{12}
Y-91	58.5 d	3.1×10^{11}	2.7×10^{12}	4.4×10^{11}	3.4×10^{12}
Pr-143	13.6 d	3.6×10^{11}	3.2×10^{12}	5.2×10^{11}	4.1×10^{12}
Nd-147	11.0 d	1.5×10^{11}	1.3×10^{12}	2.2×10^{11}	1.6×10^{12}
Cm-242	162.8 d	1.1×10^{10}	7.7×10^{10}	1.4×10^{10}	1.0×10^{11}
I-131	8.0 d	1.2×10^{16}	1.4×10^{17}	7.0×10^{15}	1.6×10^{17}
I-132	2.3 h	1.3×10^{13}	6.7×10^{06}	3.7×10^{10}	1.3×10^{13}
I-133	20.8 h	1.2×10^{16}	2.6×10^{16}	4.2×10^{15}	4.2×10^{16}
I-135	6.6 h	2.0×10^{15}	7.4×10^{13}	1.9×10^{14}	2.3×10^{15}
Sb-127	3.9 d	1.7×10^{15}	4.2×10^{15}	4.5×10^{14}	6.4×10^{15}
Sb-129	4.3 h	1.4×10^{14}	5.6×10^{10}	2.3×10^{12}	1.4×10^{14}
Mo-99	66.0 h	2.6×10^{09}	1.2×10^{09}	2.9×10^{09}	6.7×10^{09}

Source: Used with permission of the Ministry of Economy, Trade and Industry (METI).

maximum at the end of March, and it gradually decreased to 100 Bq/L in May.

The release of volatile radioactive nuclides into the atmosphere from the three units is considered to have occurred mainly after March 14, while the hydrogen explosions of units 1 and 3 occurred on March 12 and on the morning of March 14, respectively. These large releases after the night of March 14, along with the unfortunate climate conditions of wind and rain/snow at that time, have probably caused contamination over a wide region of the Fukushima Prefecture in a north-easterly direction. Along with the varying climate conditions, particularly of wind direction, some of the

released radioactivity spread in other directions, such as to the south. The dispersion of radioactive materials was tracked by the Preparatory Commission for Comprehensive Nuclear-Test-Ban-Treaty Organization (CTBTO) [8] which is a monitoring system designed to detect nuclear explosions. CTBTO reported the large-scale radiation leak resulting in the 20 km exclusion zone being set up around the power plant and people within the 20–30 km zone being advised to stay indoors.

24.2.1 Iodine-131

Iodine-131 is a fission product having a half-life of 8 days, and is important in view of its radiological risk to children by accumulation in the thyroid, which was a major issue at Chernobyl. The estimated total amount of iodine-131 released into the atmosphere lies in the range from 120 to 500 PBq, which means that there is still considerable uncertainty associated with the calculated estimate. This amount corresponds to about one fifth or one tenth of the release from Chernobyl. The latest estimate by TEPCO is about 500 PBq, while those by JAEA, NISA, and the Japan Nuclear Safety Commission (NSC) are in the range from 120 to 150 PBq. According to the study on the analysis of gaseous sample by JAEA, the release rate of iodine-131 was initially 10^{15} Bq/h, and continued to be of the order of 10^{14} Bq/h in the period from March 15 to 24. It is probable that the majority of the iodine release occurred in these ten days. Analysis of iodine deposition has been performed at 2,200 locations, and with this a map created of the radioactive contamination. This map showed that iodine-131 spread northwest of the plant, just like cesium-137 as was indicated on an earlier map. Iodine-131 was also found south of the plant at relatively high levels, even higher than those of cesium-137 in coastal areas south of the plant. According to the Ministry, clouds moving southwards apparently acquired large amounts of iodine-131 that were emitted at the time.

24.2.2 Tellurium-129m

Deposition of tellurium-129m was determined from soil samples taken during the period from June 6 to July 8 over an area of 100 km radius around the Fukushima Daiichi NPP. Tellurium-129m, which is a fission product with 33.6 days half-life is as volatile as, and shows similar behaviour to iodine. The highest concentration found was 2.66 m Bq/m^2, 2 km from the plant in the empty town of Okuma. It was pointed out that the observed ratio of tellurium-129m to cesium-137 varies with the location of the deposition. The average ratio obtained for the deposition in the north region from the NPP is about 0.19, while it was 0.88 for the coastal area in the south. In southern inland areas, it was 0.23. They may suggest that, between tellurium and

cesium, there is a different mechanism or source term, different environmental behavior, as well as a different timing of the release. The amount of tellurium-129m released is a few tenths that of iodine-131 (Table 24.3), and its radiological significance was not high compared to iodine and cesium.

24.2.3 Cesium-137

Cesium-137 is the major nuclide that causes environmental and radiological affects lasting over a long period. The estimated total amount of cesium-137 released into the atmosphere is summarized in Table 24.2 and ranges from 8.2 to 15 PBq suggesting significant uncertainty. Estimates of the released amount have been performed by several foreign institutes and researchers, e.g. [13–16]. The estimation by the French Institut de Radioprotection et de Surete Nucleaire (IRSN), which provided 30 PBq as the sum of cesium-137 and 134, agrees quite well with those of Japanese authorities. These estimates correspond to about one eighth to one tenth of the release from the Chernobyl accident. It should be noted that the distance of the cesium deposition with higher contamination than 1,000 kBq/m^2 is limited to within 80 km of the NPP in specific directions (Plate X, between pages 448 and 449), while in the case of the Chernobyl accident, more distant dispersion was observed. The release rate of cesium-137 reached 10^{15} Bq/h on March 15, but it gradually decreased to between 10^{12} and 10^{13} Bq/h, and finally to 10^{11}–10^{12}Bq/h after March 25.

The cesium-134/cesium-137 ratio observed in deposition samples over a wide area, about one month after the accident, is mostly in the range from 0.95 to 1.0, which is different from that of Chernobyl. This is expected to decrease gradually along with the decay of cesium-134 which has a half-life of 2^{-1} years.

24.2.4 Strontium-90

About 100 out of the 2,200 locations used for soil sampling were used for the analysis of strontium-89 and 90. The locations where strontium-89, having a half-life of 50.5 days, was detected is attributable to the accident, while locations where only strontium-90 was detected should be attributed to the result of weapons test fall-out made before the accident. In fact, locations without strontium-89 showed strontium-90 concentration lower than 950 Bq/m^2, which is the level of weapons testing fallout. According to the contamination map created by Japan's Ministry of Education, Culture, Sports Science and Technology (MEXT), locations where both strontium-89 and 90 were detected are distributed over an area of about 50 km radius from the NPP. In the area within about 50 km radius, the maximum concentration of strontium-90 was 5,700 Bq/m^2, but it corresponds to only 0.12 mSv

dose over 50 years, which is much lower than the effect of cesium-134 and 137. The ratio between detected strontium-89 to 90 was in the range from 1.9 to 4.0, which is in the possible range of the measurement error to be associated with the difficult detection of β-rays. On the other hand, the ratio of strontium-90 to cesium-137 was found to vary extensively from 0.00016 to 0.058 depending on the location, which suggests a non-uniform distribution of strontium compared with cesium. The ratio of radioactivity of strontium-90 to cesium-137 in the nuclear fuels in the reactor core is in the range from 0.7 to 1.0, therefore, the low ratio of strontium-90 observed for the deposition indicates the lower volatility or mobility of strontium than cesium, being the most volatile and movable element in the fuel components.

Strontium-90 of 195 Bq/kg was found in the sediment on the roof of an apartment building in the city of Yokohama, south of Tokyo, and this news caused a controversy about the possibility of long-distance transfer of strontium from Fukushima. However, it is understood to be attributable to the result of past weapons testing fall-out, and its attribution to the accident was incorrect.

24.2.5 Plutonium isotopes

The major isotopes of plutonium in the reactor fuel are plutonium-238, 239, 240, 241, and 242, which account for as high as 1% of the total weight of the heavy elements in high burn-up fuel, and is recognized to have high priority in radiological protection. Analysis of α-radioactivity of soil samples by MEXT showed non-negligible distribution of plutonium isotopes in the environment. Despite the low concentration detected in soil, plutonium was found at some locations in the region up to several tens of km radius from the NPP, especially in the northwest direction. Plutonium is generally observed in the environment as a result of weapons testing and the Nagasaki atomic bomb. The radioactivity ratio between plutonium-238 and the sum of 239 and 240 is a fingerprint for the origin of plutonium contamination in the environment. The ratio observed for the weapons testing fall-out in Japan is approximately 0.026 on average, but after the Fukushima accident it ranged from 0.33 to 2.2. This proves that the observed plutonium is from the accident.

However, according to MEXT, the sum of the radioactivity concentration of plutonium-239 and 240 was from 0.6 to 3.3 Bq/m^2, and this was within the range of the background contamination by weapons fall-out, which is 17.8 Bq/m^2 on average over the period from 2001 to 2010. Therefore, the radiological effect of the plutonium by this accident is considered to be within the existing effect of weapons testing fall-out. Zheng *et al.* [9] analysed soil samples in the area from 25 km to as far as 230 km from the NPP.

They found 1.4 Bq/kg at locations about 30 km from the NPP, which is several times higher than the background. Despite its low radiological effect, the fact that plutonium, being quite a non-volatile element, was found significant distances away, suggests the need for more careful follow-up for the environmental effects of this accident.

24.3 Dispersion and transport of radioactive materials

24.3.1 Regional dispersion

According to the May 24, 2012 press release from TEPCO, radioactivity levels of noble gases, iodine-131, cesium-134, and cesium-137 released into the air as a result of the Fukushima Daiichi NPP accident from March 12–31, 2011 were ~5×10^{17}, 5×10^{17}, 1×10^{16} and 1×10^{16} Bq, respectively [5]. Since the devices capable of directly measuring the levels of radioactive materials (such as the exhaust stack monitor) were damaged in the accident, the above-mentioned release amounts were primarily estimated through computer simulation, based on the observed air dose rates, wind direction and wind speed. These observed data were either acquired by the monitoring cars near the power station or provided by the Japan Meteorological Agency, and an over-simplified assumption of constant release rates of radioactive nuclides was often made in these simulations.

Since the beginning of the accident, radiation level measurements for a variety of environmental matrices at inland and coastal locations near Fukushima have been monitored by MEXT [10]. This comprehensive database has been serving as an important source for studies attempting to simulate the dispersion and transport of the released radionuclides. Starting on March 11, 2011, IRSN participated in the analysis of developments and probable radiological consequences of the Fukushima Daiichi NPP accident. With the calibration by local monitoring data, IRSN used the three-dimensional *ldX* model of its C^3X platform to model the atmospheric dispersion of released radionuclides at the regional level (several hundred to several thousand km) and reconstructed the release history of radioactive materials as follows [11]. The first releases, occurring between March 12 and 14, 2011, spread mainly northward, then northeast and east, over the Pacific Ocean. On March 15 and 16, the radioactive releases from unit 2 spread over Japan, but the weather conditions were changing rapidly. On March 16 and the following days, the releases spread eastward, moving over the Pacific and sparing most of Japan. Between the afternoon of March 20 and 23, the radioactive releases again spread over Japan. After March 23, the contaminated air masses moved toward the Pacific. The subsequent releases have been too low to cause a significant increase in the radioactivity in Japan's terrestrial environment.

During the spread of the contaminated air, a portion of the airborne radionuclides in the form of very fine particles (aerosols) or soluble gases (such as the portion of radioactive iodines) were deposited on the ground either in the form of dry deposition or wet deposition. Dry deposition occurs when the radioactive plume directly or indirectly (by first incorporating into dust or smoke) falls to the ground, while wet deposition means that the radioactive plume combines with water droplets or snow first before falling to the ground. Both types of deposition contributed to the spread of radionuclides over the terrestrial environment of Japan. Similar deposition mechanisms also occurred in areas outside of Japan when the contaminated air dispersed over the world. A unique situation presented by the Fukushima accident is the overlap of a release phase with a fallout phase beginning on March 16 [11]. The release phase lasted for 12 days starting on March 12 with a threat of new releases lasting for at least several more weeks. In addition to the immediate risk of exposure to the radioactive plume in the release phase, the fallout phase also became significant after March 16 owing to the fallout from the first atmospheric contamination event.

To understand the widespread effects of contamination by radioactive material, and to assess doses and the deposition of radioactive materials for future evacuation zones, the MEXT and US Department of Energy (US DOE) jointly performed airborne monitoring, checking the air dose rate 1 m above the ground surface within 80 km of Fukushima Daiichi NPP and the deposition of radioactive materials on the ground surface [12]. The map of air dose rates at 1 m from the ground surface measured between April 6 and 29, 2011, is shown in Plate X (a) (between pages 448 and 449), which represents the radioactive dispersion around Fukushima. After both dry and wet deposition of contaminated air, the total deposition of radioactive cesium-137 and cesium-134 in the soil surface (Plate X (b)) were found to be similar to the radioactive dispersion.

The radioactive materials in the air not only polluted the terrestrial ground surface, but also contaminated the surface waters dozens of kilometres from the NPP. This is probably the main source of the observed radioactive pollution in seawater (cesium-137 and iodine-131 concentrations of 2–27 Bq/L and 3–57 Bq/L, respectively) 30 km offshore from the damaged power plant before March 30, 2011 [11]. However, in addition to the airborne spreading mechanism, more serious pollution was caused by the leaking of water which was used to cool the damaged reactor. In particular, a crack in the pit adjacent to the unit 2 turbine hall led to the release of heavily polluted water directly into the sea. On April 6, 2011, at approximately 6 a.m. local time, TEPCO successfully stopped this release by plugging the leak with an injection of sodium silicate. Current estimates of direct marine release are usually based on the quantification of this leakage incident.

Since the half-life of cesium-137 is much longer than that of iodine-131, after the serious radiation pollution of the seawater, the concentration of cesium-137 in the seawater was measured, and its spatial distribution between April 11 and July 11, 2011 was modeled by IRSN, as shown in Plate XI (between pages 448 and 449). The concentration of cesium-137 fell sharply with time. Between April 11 and 18, the seawater outlet point of the damaged nuclear plant had a cesium-137 concentration around 900 Bq/L, which by the following week was significantly reduced to around 200 Bq/L. At the same time, the area with concentrations above the detection limit (around 5 Bq/L), the coloured zones shown in Plate XI, also decreased.

24.3.2 Global dispersion and transport

The spread of radioactive pollutants was not confined to Japan. Due to the prevailing westerlies during the accident, the radioactive nuclides had the potential to be transported offshore, across the Pacific Ocean, and further to the North American continent. Monitoring of seawater, soil and atmosphere was being done at 25 locations on the plant site, 12 locations on the boundary, and other locations further afield [1]. Trace amounts of radiation, including iodine-131, cesium-134 and cesium-137, were being observed around the world (New York State, Alaska, Hawaii, Oregon, California, Montreal, and Austria) [13]. Radioactive isotopes originating from Fukushima were picked up by over 40 CTBTO radionuclide monitoring stations [15].

On March 17–18, 2011, the first arrival of the airborne fission products, iodine-131, iodine-132, tellurium-132, cesium-134, and cesium-137, was detected in Seattle, Washington (USA) by their characteristic gamma rays. Leon *et al*. [16] used a NOAA HYSPLIT model to assess their transport time and possible trajectories across the Pacific. Plate XII (between pages 448 and 449) shows three trajectories of the radioactive nuclides, which indicate the range of transport pathways. The start time was set to March 12, 2011 at 10 UTC (Coordinated Universal Time), which was approximately 3 hours after the reported explosion from unit 1. The trajectories were calculated for three heights in the atmospheric boundary layer: 500, 1,000, and 1,500 m above ground level. The 500 m trajectory was found to be caught up in and raised by a cyclonic system over the Bering Sea. The trajectories started at 1,000 m and 1,500 m were also partially lofted but did not get involved in the cyclonic pattern. Instead, they were found to be rapidly transported across the Pacific. Upon arrival at the west coast of the US, the transport again split, with one arm transported to the north in a cyclonic direction around a low pressure system located off the coast of Washington state. There were rain showers and cool weather in western Washington at the arrival time of the plume, and the strong divergence and

precipitation associated with these weather systems most likely significantly reduced the concentrations of radionuclides that were transported. The trajectory initially started at 1,500 m was transported in the boundary layer towards California. Overall, the trajectories support the notion of transport of the radionuclides from the Japanese boundary layer to the US boundary layer in only 5–6 days. This result is significantly faster than the other previous work which examined the trans-Pacific transport times [17], especially considering the radionuclides were released in the boundary layer over Japan and measured in the boundary layer along the US west coast.

After crossing the North American continent, the contaminated air masses were anticipated to continue to move towards the North Atlantic and reach Europe. The first sign of the radioactive material in Europe was detected on March 19, 2011 in Iceland, 7 days after the explosion of the unit 1 reactor. On March 23–24, most European countries had detected the radiation. Around March 28–30, the first radioactive peak was observed. The time- and spatially-averaged values from March 20 to April 13 in Europe were 0.076 and 0.072 mBq/m^3 for cesium-137 and cesium-134, respectively [18]. Cesium-137 airborne activity levels reported after the Fukushima Daiichi NPP incident were at least 10,000 to 100,000 times lower than those observed after the Chernobyl accident. Regardless of the radionuclide considered, airborne activity levels remained sufficiently low as to be of no concern to public health in Europe. Although the prevailing wind during the accident was westerly, the radiation effect in Hong Kong, more than 2,000 miles southeast from Fukushima, was also detected before April 14, 2011. An activity of iodine-131 of 62.5 μBq/m^3 was first detected on March 26, 2011, and a maximum value of 828 μBq/m^3 was observed on March 29, 2011, in Hong Kong [19].

24.3.3 Forecast of Fukushima radioactive contamination over the next 20 years

On December 16, 2011, TEPCO confirmed that the release of radioactive materials was under control and that radiation doses were being significantly reduced [5]. In April 2012, the predicted equivalent radiation doses per year for areas near the Fukushima Daiichi NPP for the next 20 years were released [20]. As shown in Plate XIII (between pages 448 and 449), a dose of more than 100 mSv/yr may still be encountered about 23 km northwest of the plant until March 2013, and the 50 mS/yr dose zone can only be confined to a 20 km radius after March 2017. Since the standard worker dose limit for Japanese workers is 50 mSv per year and 100 mSv over 5 years [21], certain areas will still be subject to high alert for radiation effects in the near future. By March 2022 (11 years after the accident), certain hot spots may still possess a radiation dose higher than 50 mS/yr. These hot

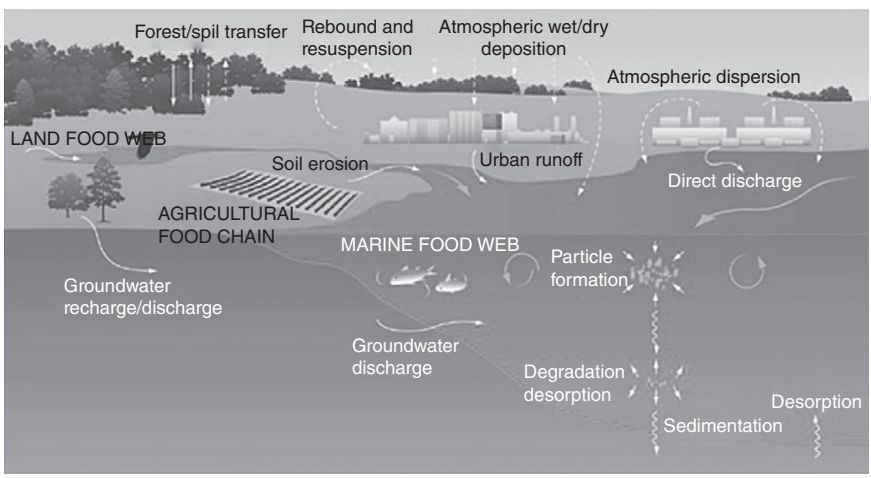
spots will mostly be eliminated after 20 years, as shown in the prediction for March 2032 in Plate XIII.

24.4 Effects of released radiation on food, environment and human health

The Fukushima NPP accident resulted in the spread of radionuclides into the atmosphere. The radionuclides were volatilized by the high temperature in the reactor core and during the explosions and fires. In addition, seawater containing non-volatile activation products and fuel rod materials may have been released into the subsurface and ocean environment [22]. The fate and potential transport mechanisms of these radioactive materials are shown in Fig. 24.2; the illustrated atmospheric, terrestrial, and aquatic systems were all affected by the accident. Since some long-lived radionuclides were among the released radioactive materials, the radioactive contaminants may have a profound impact on the environment, food, and human health through their migration between and within these systems.

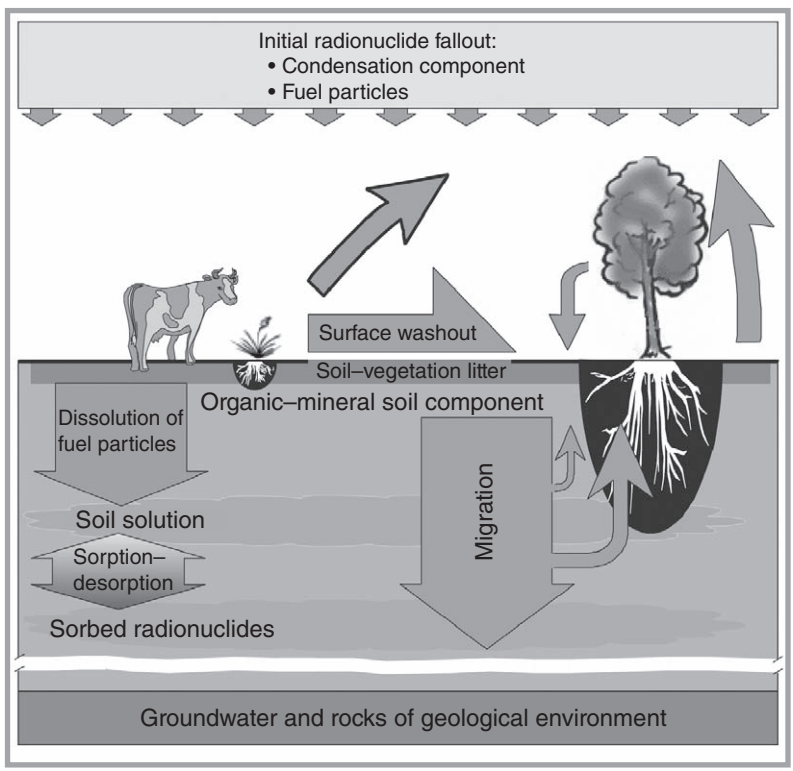
24.4.1 Environmental impact

Radioactive noble gases (e.g., krypton and xenon) and volatile fission products (e.g., iodine and cesium) were the main constituents of the radioactive materials released into the atmosphere during the Fukushima accident [24]. Of all the noble gases, krypton-85 has the longest half-life (10.8 years) and



24.2 Potential migration of radionuclides in atmospheric, terrestrial, and aquatic systems [23]. Used with permission from The American Association for the Advancement of Science.

will remain in the atmosphere for a very long time due to its high chemical stability. In general, the inhalation of krypton-85 by animals or humans adversely affects the organs of the respiratory system. However, taking into account that we are naturally exposed to much extensive inhalation of radon and daughter nuclides, and that a considerable amount of krypton-85 has already been accumulated in the air by artificial activity, the effect of krypton-85 by this accident is considered to be limited. Radioactive materials containing iodine, cesium and other radionuclides are often carried by air particles and subsequently introduced by wet and dry deposition into the terrestrial environment. Radionuclides behave differently in the terrestrial environment. Some of them (such as cesium and iodine) are mobile in the environment and can easily be transferred into the water supply and food chain [25]. Other radionuclides have low solubility (such as the actinides) and can largely be retained in the soil [26]. The main transfer pathways of radionuclides in the terrestrial system are shown in Fig. 24.3.



24.3 Main transfer pathways of radionuclides in the terrestrial system [27]. Used with permission from Springer.

The marine environment was affected by aerosols emitted into the atmosphere and then deposited on the ocean, as well as by the direct release of seawater used for cooling the reactors. In the future, leaching from contaminated soils will be the main source of pollution into the marine environment. The mobility of radionuclides transferring between atmospheric, terrestrial, and aquatic systems increases the scope of their adverse influences to a wide variety of living organisms and ecological processes.

24.4.2 Impact on foods

Radioactive materials released from the Fukushima NPP have contaminated leaves of plants exposed to the air and also is very likely to be in the stems of plants adsorbing nutrients from the contaminated soil. As a result, radioactive materials may enter the food chain for human consumption. Between mid-March 2011 and February 8, 2012, three categories of foods were sampled to check for radiation contamination: plant-based foods (e.g., vegetables, tree fruits, bamboo shoots, tea leaves, rice and other cereals), animal-based foods (e.g., cow's milk and meat), and foods from natural and semi-natural environments (e.g., forest products and aquatic species). These tests included 104,318 food samples from different sites in Japan (not including Fukushima), and about 1% of these samples showed signs of contamination exceeding the standard limits for sale or consumption in Japan. In the Fukushima area, 18,350 samples were examined, and 3.5% of them were determined to exceed the standard limits [11].

24.4.3 Impact on human health

The International Atomic Energy Agency (IAEA) [28] and the World Health Organization [29] indicate that radiation exposure can result in both short-term and long-term effects in every body organ. In the Fukushima accident, public concern focused on both acute radiation sickness and increased long-term cancer risk [30].

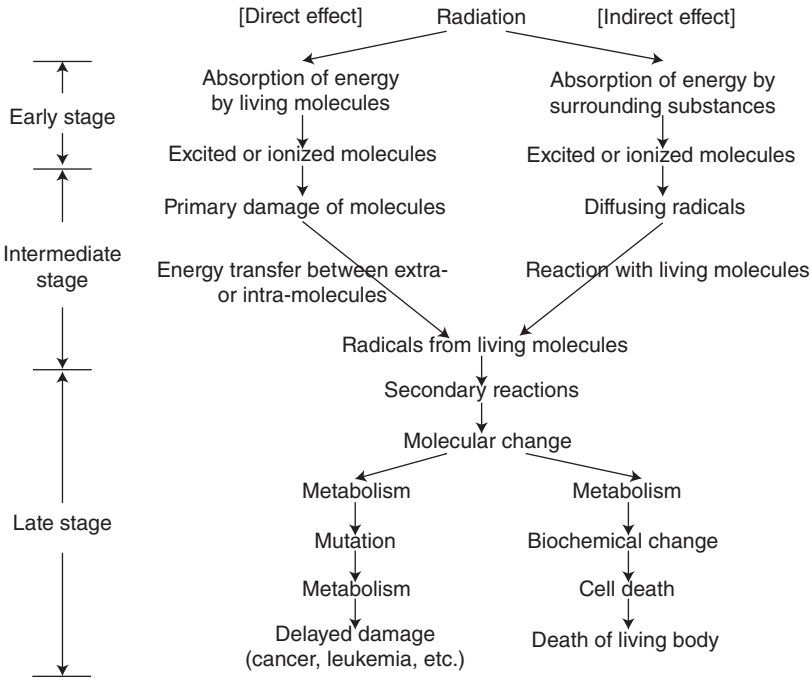
Three months after the Fukushima accident, 20 teams were dispatched to Fukushima to screen for human radiation exposure. More than 5,000 people were screened by the staff of Hirosaki University (Fig. 24.4), and the results showed no acute radiation injuries [31]. The six deaths associated with the operation of the NPP were not attributable to exposure to ionizing radiation [11]. However, biological responses after exposure to radiation are time-dependent (Fig. 24.5), and the lack of symptoms appearing in the short term does not indicate freedom from long-term adverse effects due to radiation exposure. In the case of the Chernobyl accident, although no cases of cancer were confirmed to be due to the radiation exposure, some studies suggest that the risk of thyroid cancer for children living in nearby

24.4 Screening for human radiation exposure carried out by Hirosaki University staff [31]. Used with permission from the Institute of Applied Biochemistry.

areas could have been increased by a factor of 2 to 5 per 1 Gy of thyroid dose [30]. The Japanese government invited children exposed to the radioactive releases at different districts to be evaluated for thyroid disorders. As at December 31, 2011, 14,442 children had undergone screening, and no cases of fluid-filled cysts larger than 20 mm in diameter were found. Between January and March 2012, 27,467 more children were screened for thyroid. Furthermore, starting in April 2014, a follow-up thyroid screening will be performed on 360,000 potentially affected children once every two years until age 20 and every five years above the age of 20 [11]. No information is currently available concerning the progress of the studies planned for pregnant women and evacuees.

24.5 Clean-up programme

Although the release of radioactive materials from the damaged PCVs is now under control and the public's radiation exposure from additional releases has been significantly diminished (<1 mSv/yr) [32], the explosion and radiation emission from the Fukushima accident results in a need to clean up the problematic wastes, a process that will take many years. This



24.5 Time-dependence of biological responses after exposure to radiation [31]. Used with permission from the Institute of Applied Biochemistry.

task will need to include the treatment of a huge amount of accumulated water used for cooling the damaged SF pools and cleaning contaminated debris, surface soil, vegetation, structures surrounding the SF rods, as well as the sludge derived from processing the water potentially containing radioactive materials. The clean-up programme has been initiated. As required by the Japanese government, a mid- and long-term roadmap was drafted by TEPCO, the Agency for Natural Resources and Energy (ANRE) and NISA. This roadmap was finalized at the government and TEPCO's mid- and long-term countermeasures conference on December 21, 2011. The four basic policies addressing the mid- and long-term issues are the safety of local citizens and workers, which was given a top priority; the maintenance of transparent communications with local and national citizens; a continuously updated roadmap based on the on-site situation and the latest R&D results; and coordination efforts for TEPCO, ANRE, and NISA to achieve the clean-up goals [33].

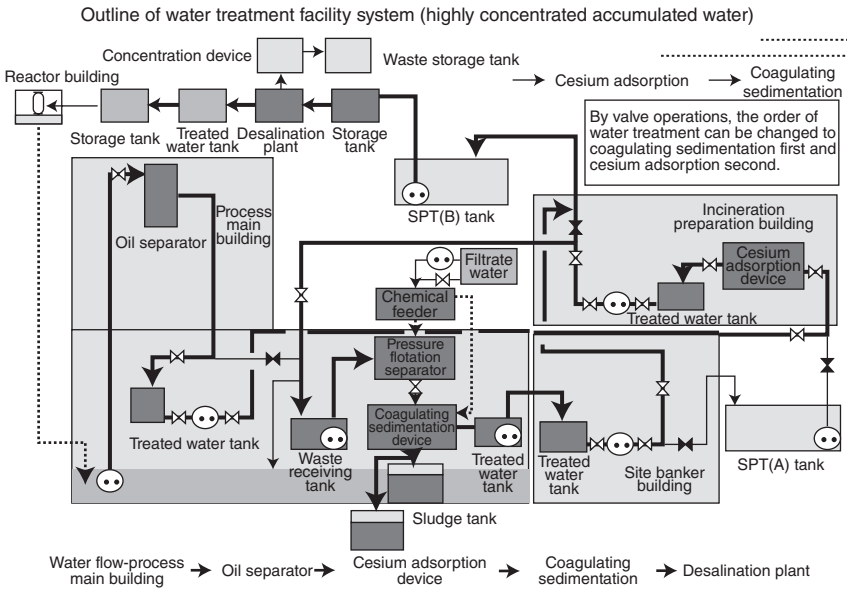
In the Fukushima accident, around 56% of the total radioactive materials released into the atmosphere were noble gases [5]. The released gases chiefly consisted of xenon-131, which has a half-life of only 5.2 d, and most

of the gaseous emissions occurred in the early days of the accident. Therefore, the radioactive gases would have decayed, spread and been diluted to a very low radiation level. Gas pollutants are thus not assumed to be a threat to humans and the ecosystem and are not a focus of the clean-up programme. Merely as a preventive measure, PCV gas control systems were installed and are still operating at units 1, 2, and 3 to avoid any potential further emissions [32].

Treatment of the cooling water is clearly a bigger challenge than the gaseous waste processing in the clean-up programme. Firstly, the cooling water which was in direct contact with the damaged fuel rods was seriously contaminated by a variety of nuclides. Secondly, the injection of seawater into the damaged reactors not only promoted corrosion but also impeded the usual water treatment processes. The higher sodium concentration (ionic strength) of seawater reduces the specific adsorption of cesium when using zeolite to adsorb pollutants from water. As a result, more zeolite is required, and more spent zeolite waste will be produced. Before the commencement of operations of two water treatment systems in June 2011, a huge volume of contaminated water had been accumulated in surface storage tanks [21]. If water leakage had occurred into the RPV, the overall water treatment task would have been further burdened. In spite of these challenges, nearly 90% of the treatment system capacity was achieved by mid-August 2011 [21]. The Fukushima Daiichi nuclear power station now operates a large water treatment facility as shown in Fig. 24.6. As of July 3, 2012, the water treatment capacity had reached 540 m^3/d, and the total volume of water treated to date had reached 196,091 m^3. The treated water has been circulated into units 1, 2 and 3 for reactor building decontamination at rates of 132 m^3/d, 204 m^3/d, and 204 m^3/d, respectively [34].

The treatment system (Fig. 24.6) comprises four parts [35]:

1. an oil separator,
2. a Cs-adsorption system developed by the US company Kurion, which itself consists of three parts (a pre-treatment column packed with a surfactant modified zeolite aluminosilicate sorbent for removing remaining oil and Tc, four parallel columns of the sorbent herschelite for removing Cs, and a column packed with Ag-impregnated herschelite sorbent to remove I). These filters are porous zeolites that loosely bind metal ions and through a combination of adsorption and ion exchange trap Sr^{90}, Cs^{134} and Cs^{137},
3. a system for removing the remnant Cs provided by the French company Areva which uses precipitation and coagulation,
4. on August 19, 2012, a second line called Sarry developed by Toshiba-Shaw, was added in parallel to the Kurion-Areva line. This line uses Cs adsorption by crystalline silico-titanates (CST).



24.6 Water treatment scheme used in the Fukushima Daiichi NPP to remove radioactive contaminants in the water [21]. Used with permission from Tokyo Electric Power Company (TEPCO).

The decontaminated water goes to tanks where it mixes with reagents such as nickel ferrocyanide and barium sulphate, along with polymers and sand. The dissolved radioactive metals form precipitates and colloids, which are trapped as a radioactive sludge, allowing the water to be desalinated by reverse osmosis and by evaporation, these desalination processes were added on June 24 and August 7, 2012, respectively, in a shielded ion-exchange module. The two processes reduce the concentration of cesium – the major element of concern from the reactors – in the water by up to a million times [35,36].

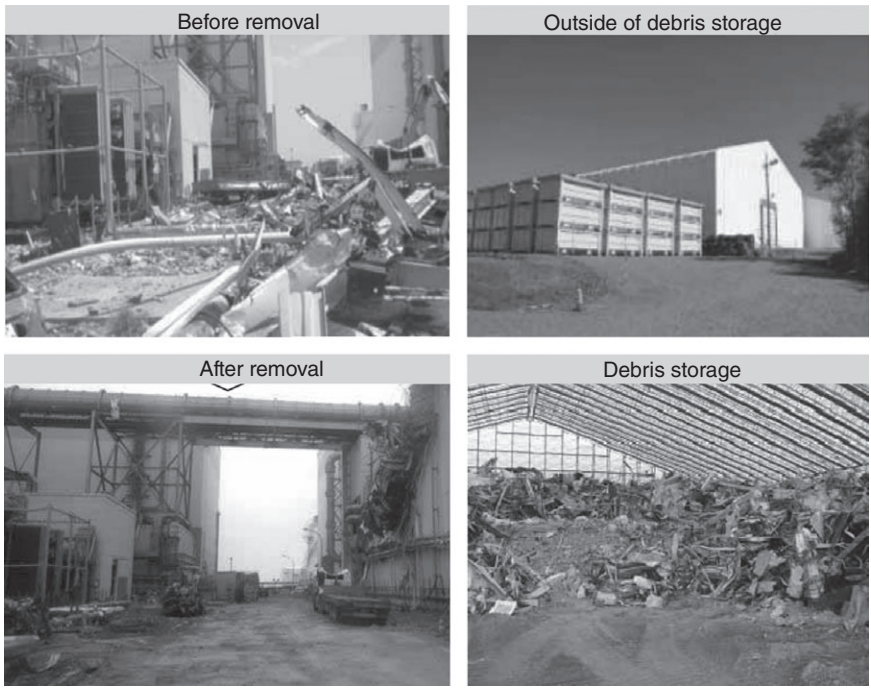
A problem still remaining in the water treatment task is the need for water storage. The American Nuclear Society (ANS) pointed out a contribution of 200–500 m^3/day of contaminated water from groundwater in-leakage, but TEPCO is unable to release this water due to existing environmental policy [21]. Furthermore, a large volume of tritiated water, with a tritium concentration of 1,000 Bq/m^3, also needs storage, since the half-life of the tritium is about 12 years. Generally, the daily accumulation of water is about 200–700 m^3, and the need for water storage will eventually challenge the existing storage capacity, even with the new additions of >111,000 m^3 of tanks and 10,000 m^3 of megafloat barges. Currently, the water storage operations, as well as the forecast conditions, are required to be submitted to the NISA weekly, providing oversight to the water processing [34].

After sealing the crack in the pit adjacent to the reactor 2 turbine hall and terminating the discharge of highly radioactive water, TEPCO took action to mitigate the level of radioactivity in the contaminated ocean. On April 16, 2011, TEPCO dumped about 10 bags of zeolite in the seawater area near the Fukushima Daiichi NPP, as shown in Fig. 24.7. Each bag contained 100 kg of ground zeolite and would be raised periodically to check the radiation level [37, 38].

The secondary solid waste (sludge, spent zeolite, and the used reverse osmosis membranes) generated from the water treatment operation are stored in the plant and labeled as radioactive solid waste. Treatment and immobilization of these secondary wastes is now an issue. One option being examined for the zeolite wastes is use of a mobile vitrification system using a form of in-can melting. The Areva process creates a sludge and the CST IE-911 inorganic Ti-based resin is also being used. No decision has yet been made on what to do with the Si and Ti spent resins but cementation and vitrification are being considered.

Currently, there is no clear plan for the treatment of such secondary waste, although the need for R&D in cementation and disposal techniques is mentioned in the mid and long-term roadmap published by TEPCO [33].

24.7 Submerging zeolite into a water outlet from the Fukushima Daiichi NPP to reduce the radiation contamination to the ocean environment [39]. Used with permission from Tokyo Electric Power Company (TEPCO).



24.8 Debris waste generated from the Fukushima Daiichi nuclear plant incident and its temporary storage [40]. Used with permission from Tokyo Electric Power Company (TEPCO).

In addition to the secondary solid waste, approximately 28,000 m^3 of debris on the plant site from the disaster itself has already been collected by remote-controlled vehicles. These solid wastes were classified according to their material types as well as their level of radiation [32]. About 6,000 m^3 of debris from the yard area around the nuclear plant is stored in ~900 metal containers (with a volume of 4 or 8 m^3 per container) and will be transported for off-site treatment [21, 32]. Larger and less contaminated items are stored in bulk in a new solid waste building, pictured in Fig. 24.8. Before decontaminating the RAW, characterization and compositional analysis of the stored debris are required. The debris waste with the strongest radiation will be from the damaged reactors as well as in the damaged SF pools.

24.6 Mid- and long-term roadmap

With stable conditions in the reactors of the Fukushima Daiichi NPP achieved, TEPCO will continue to monitor the radiation levels in the plant and carry out mid- and long-term work to achieve the following targets [33]:

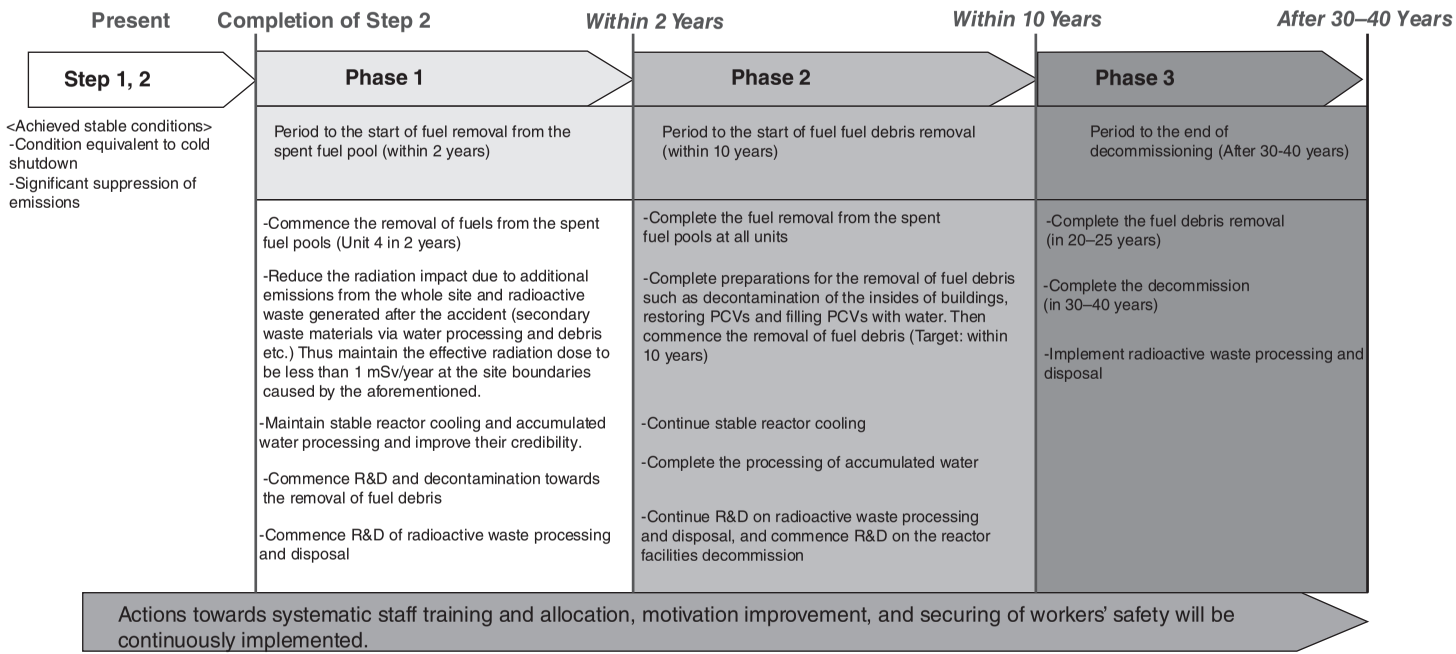
1. In 2012, processing facilities for multi-radioactive nuclides, which could not be removed by the current cesium adsorption units, were installed. With the sealing of the water leakage, the processing of the accumulated water will be accomplished within 10 years.
2. To mitigate seawater contamination, seawater purification continued to be operated until the end of 2012. Furthermore, a water shielding wall will be installed by 2014 to prevent local groundwater discharge into the ocean.
3. Removal of fuel from SF pools will commence within 2 years, in the sequence of units 4, 3, 1, and 2. Complete fuel removal for all units will take about 10 years. Fuel reprocessing and storing methods will also be studied during this period.
4. After fuel removal, the leftover fuel debris will also be removed in accordance with site conditions, safety requirements, and the development of remote control technologies. This removal will be initiated within 10 years and is expected to be completed after 20–25 years.
5. The reactor facilities of units 1–4 will also be demolished within 30 and 40 years.
6. Another significant target is the processing and disposal of the radioactive waste. By the end of 2012, an R&D plan for the post-accident waste was set up. The R&D programme includes, but is not limited to, waste identification, effective waste treatment and disposal methods, potential equipment/device development. At this stage, the complete disposal of the RAW is expected to be accomplished after 30–40 years.

The overall mid- and long-term roadmap published by TEPCO is summarized in Fig. 24.9.

Options for managing the corium product of the melted cores at Fukushima have been considered [6] incuding stabilizing the site by creating a protective sarcophagus as is being done at Chernobyl. However, an understanding of the interaction between water and the corium with which it would undoubtedly come into contact must be developed if this option is eventually chosen.

24.7 Sources of further information

A number of publications about Fukushima are now becoming available including a special issue of *Elements*, the international magazine of mineralogy, geochemistry and petrology (June 2012) with articles on the resulting atmospheric dispersion, land contamination and oceanic dispersion simulations. The Kyoto University Research Reactor Institute recently published the proceedings of an *International Symposium on Environmental Monitoring and Dose Estimation of Residents After Accident of TEPCO's*



24.9 Summary of the mid- and long-term roadmap for waste clean-up and decommissioning of the Fukushima Daiichi NPP [33]. Used with permission from Tokyo Electric Power Company (TEPCO).

Fukushima Daiichi Nuclear Power Stations as part of the KUR research program for scientific basis of nuclear safety.

24.8 Acknowledgements

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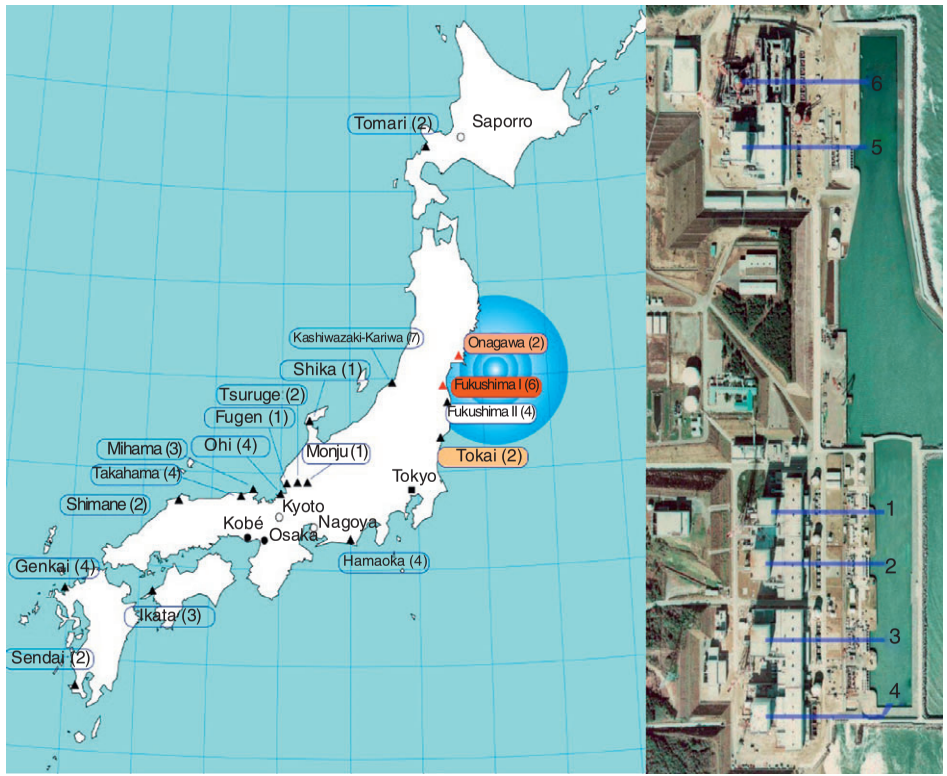
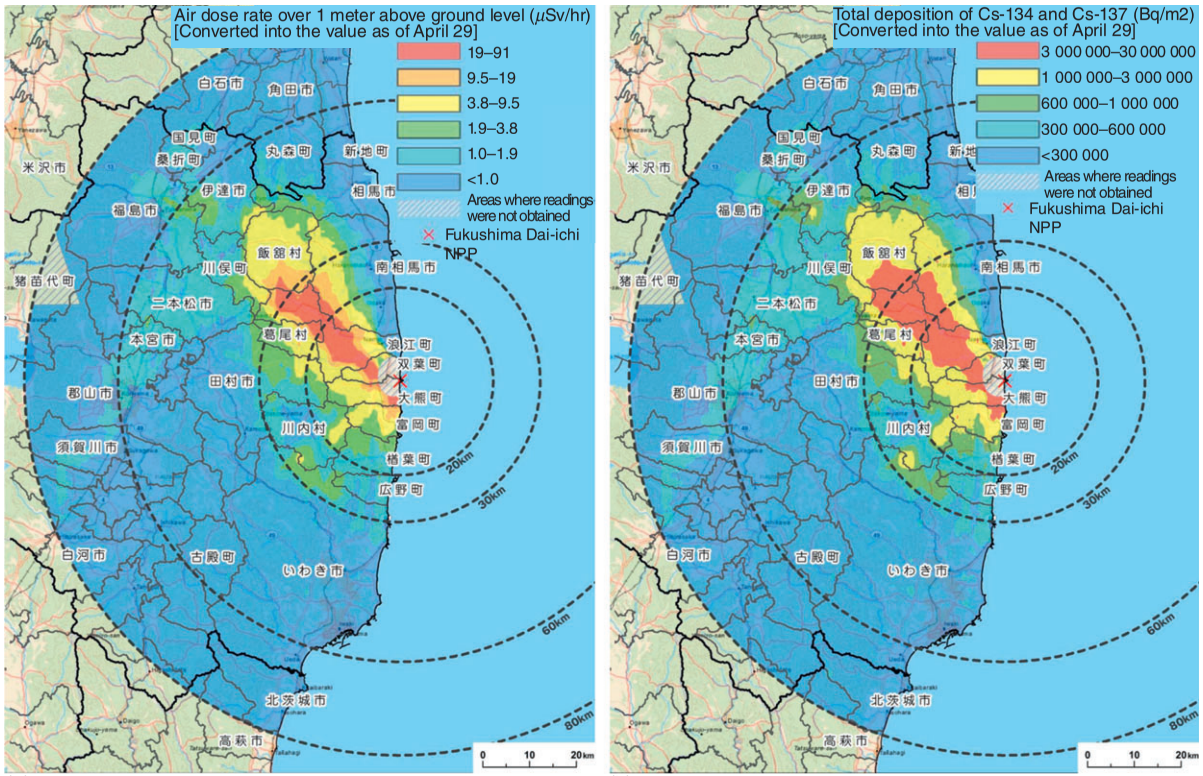


Plate IX (Chapter 24) Fukushima Daiichi NPP, based on national land image information. (Left), The number in parentheses represents the number of reactor units on every site. (Right), location of reactors 1–4 and site for reactors 5 and 6; north is up (Color Aerial Photographs).



(a) (b)

Plate X (Chapter 24) The radioactive dose data of (a) air and (b) total surface deposition of cesium-134 and cesium-137 inside the 80-km zone of the Fukushima Daiichi NPP [12].

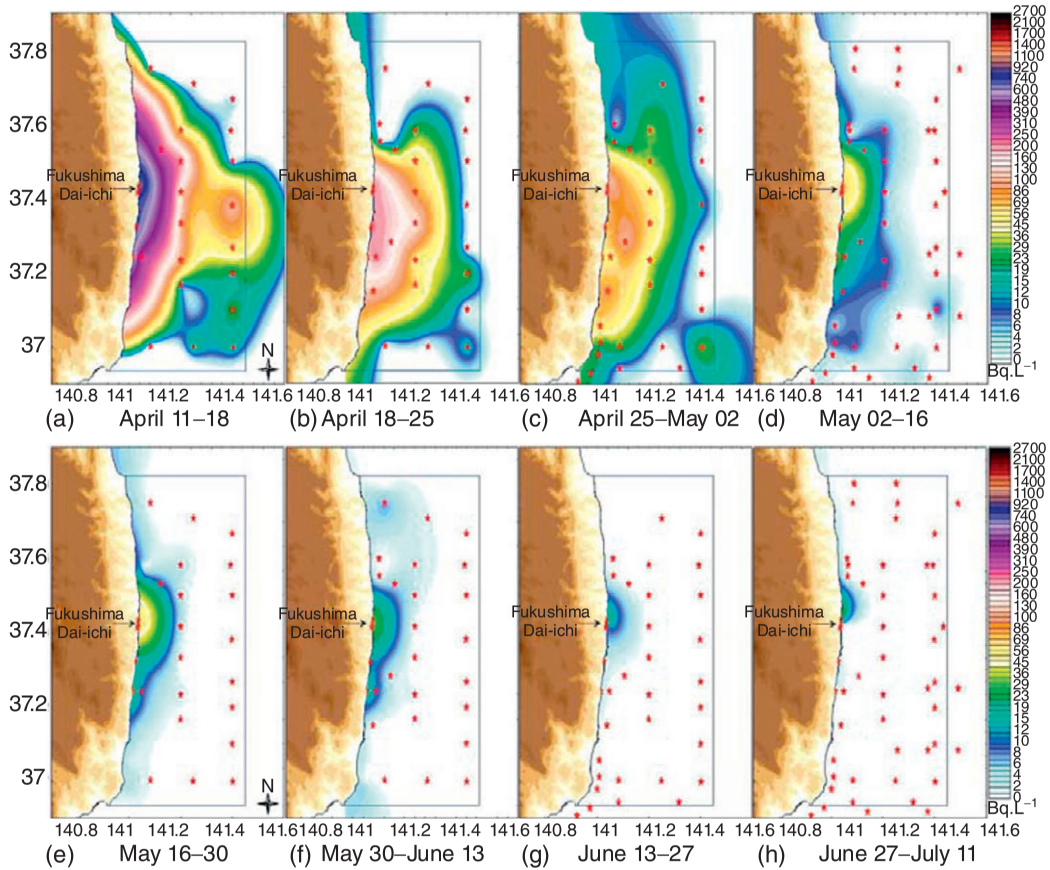


Plate XI (Chapter 24) (a–h) Spatial distribution of cesium-137 concentration in the seawater observed between 11 April and 11 July 2011 near the Fukushima Daiichi NPP. The red points indicate sampling points [11].

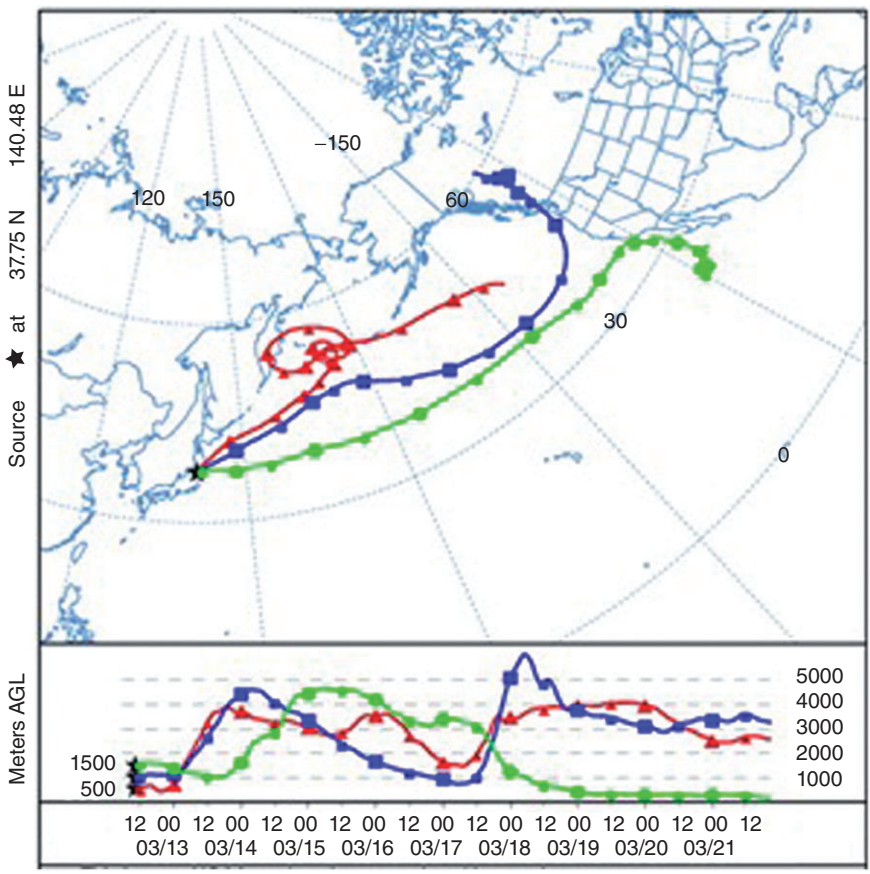


Plate XII (Chapter 24) Calculated trajectories for radioactive nuclides released from the first explosion of the unit 1 reactor of the Fukushima Daiichi NPP [16].

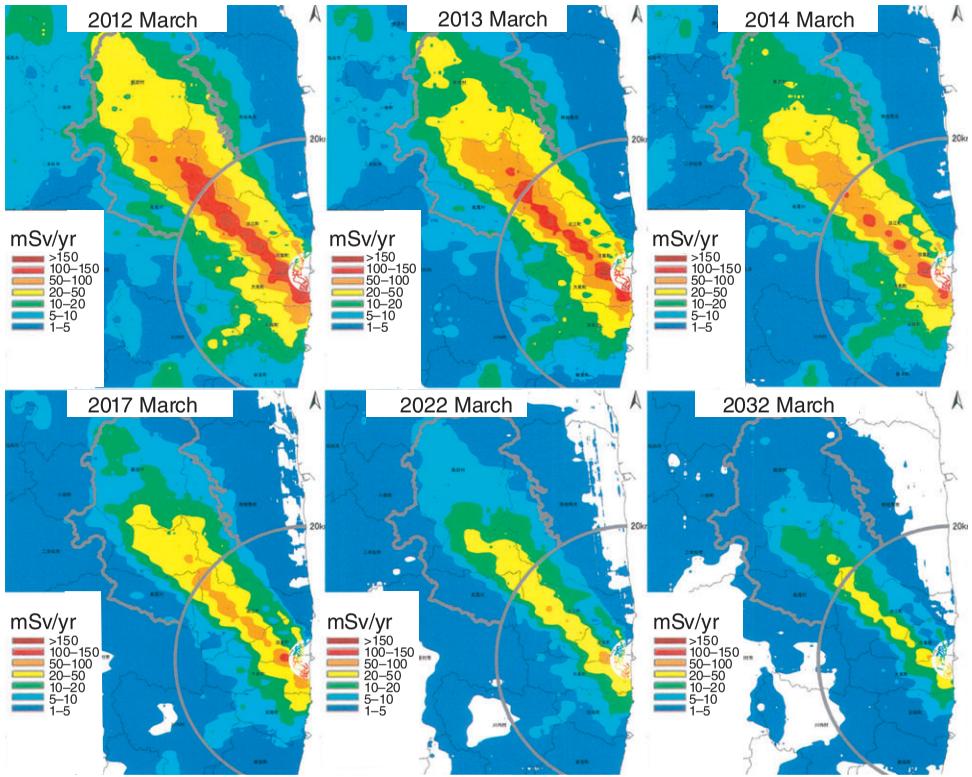


Plate XIII (Chapter 24) The predicted radioactive dose map of the Fukushima Daiichi NPP and its surrounding areas in 2012, 2013, 2014, 2017, 2022 and 2032 [20].

25

Management of radioactive waste (RAW) from nuclear weapons programmes

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Abstract: Although small quantities of radioactive waste existed before nuclear weapons were built, their manufacture led to a dramatic increase in both the quantity of waste and its activity. These wastes arose from the entire manufacturing process starting with the supply of fissile material, the production of warheads and, more recently, the excess of fissile material stocks generated by the reduction in warhead numbers by the major powers. When compared with civilian nuclear wastes, defence wastes can offer different problems related to their disposal which require specific treatment, immobilization or disposition techniques to be developed. There is no universal agreement on the ultimate disposal techniques as these are dependent on several factors both scientific and political, although the most likely option will probably involve geological disposal of immobilized, passively safe waste forms in underground repositories. Immobilization may involve vitrification or incorporation into a ceramic host. In this chapter we discuss the approaches taken by several countries to address the treatment and disposal of defence wastes or excess material.

Key words: nuclear wastes, immobilization, plutonium, highly enriched uranium (HEU), pyrochemical wastes, borosilicate glass, synroc-type ceramics, high level waste, transuranic waste (TRU), waste form, MOX, waste oil, geological disposal, non-proliferation.

25.1 Introduction

Radioactive waste existed long before the existence of nuclear weapons (e.g., the use of radium paints for luminous dials), but it was the advent of the US Manhattan Project during the Second World War that greatly increased both the quantity and activity levels being produced. This in turn required an engineered solution for the waste generated rather than the less formal disposal methods prevalent at the time, which included deep sea disposal. Therefore storage rather than disposal was the solution imposed by the exigency of war. The subsequent proliferation of nuclear weapons programmes in the late 1940s and 1950s, coupled with the ever increasing number of warheads, created a vast legacy of wastes arising from the production of Pu and U metals together with tritium, and their manufacture

into warhead components. At one US site alone, Hanford in Washington State, production of radioactive materials for the US nuclear weapons programme generated 53 million gallons of liquid waste which was stored in 177 carbon steel tanks buried in the ground (Bearden and Andrews, 2007). These acidic wastes were neutralized by the addition of sodium hydroxide in order to minimize corrosion of the storage tanks. The majority of this waste arose from the reprocessing of reactor fuel elements to generate plutonium, rather than resulting from the direct warhead manufacturing process. Although this waste can be ascribed to nuclear weapons programmes, it is also legitimate for it to be described as site remediation, as over the decades the storage tanks have deteriorated to the extent of allowing some radioactive and corrosive liquids to leak into the soil and groundwater. In this chapter we shall discuss progress on immobilizing these wastes, but will concentrate on those wastes which can be more directly ascribed to the production of nuclear weapons.

In addition to the United States, there are four other countries covered under the Non-Proliferation Treaty (NPT) which are described as nuclear weapons states that have detonated a nuclear device prior to January 1967 (see http://www.un.org/disarmament/WMD/Nuclear/NPT.shtml). These are Russia (the former USSR), the United Kingdom, France and China. These countries are officially recognized under the NPT as possessing nuclear weapons. Three other states, India, Pakistan and Israel, are known to possess nuclear devices but are not signatories to the NPT. Other countries known at one time to have had active nuclear weapons programmes include South Africa, Argentina, Brazil, Iraq and Libya, but these states cancelled programmes before obtaining useable devices. Currently, North Korea (which withdrew from the NPT in 2003) is known to be pursuing a nuclear weapons programme, whilst Iran and Syria are thought to be. All these countries have, or have had, radioactive wastes from their respective weapons programmes requiring attention.

25.2 Waste types, classification and composition

25.2.1 Waste classification

Radioactive wastes were originally classified into high, medium and low level, but as the nuclear industry has progressed, additional categories have been introduced and some have been sub-divided (Table 25.1). Having well-defined classifications is important, as these frequently form the basis on which national governments base their legislation relating to the disposal routes for radioactive wastes.

Several waste categories are clearly defined by their activity levels based on either the α or β/γ activity. High level waste is defined by its

Table 25.1 Waste definitions

Category	Typical characteristics
Low volume very low level waste (VLLW)	Contains $< 4 \times 10^5$ Bqte^{-1} (β/γ)
High volume very low level waste (VLLW)	Contains $< 4 \times 10^6$ Bqte^{-1} (β/γ)
Low level waste (LLW)	Contains $< 4 \times 10^9$ Bqte^{-1} (α); $< 12 \times 10^9 < 12 \times 10^9$ Bqte^{-1} (β/γ)
Intermediate level waste (ILW)	Contains $> 4 \times 10^9$ Bqte^{-1} (α) or $> 12 \times 10^9$ Bqte^{-1} (β/γ) but for which its heat output need not be taken into account in the design of waste storage or disposal facilities
High level waste (HLW)	Radioactive waste in which the temperature may rise significantly as a result of its radioactive content, so that this factor has to be taken into account in the design of waste storage or disposal facilities
Transuranic waste (TRU)	>100 nCig^{-1} (α) from transuranic elements with half-lives >20 years.

Source: UK Committee on Radioactive Waste Management (2011).

heat-generating ability and the IAEA have recently revised their definition by removing the 2 kWm^{-3} threshold (IAEA, 2009).

In the UK, low volume VLLW can be disposed of safely to unspecified destinations with municipal, commercial or industrial waste, whereas high volume VLLW can only be disposed of to a specified landfill site. For wastes containing solely tritium or carbon-14, the limits are increased by an order of magnitude.

Wastes arising from nuclear weapons programmes can fall into all of the above categories, but the three of specific interest are HLW, TRU and ILW. In the early days, many of the candidates investigated for the immobilization of commercial wastes were considered, but currently ILW is generally compacted and cemented into steel drums, whilst HLW and some ILWs are vitrified in borosilicate glass; however, some of the newer wastes may require alternative immobilization techniques to be developed. Disposal routes already exist for LLW, which for the UK is in a special site at Drigg in Cumbria.

25.2.2 Comparison of commercial and defence wastes

Commercial wastes are produced from a variety of sources but derive predominantly through the generation of electricity using nuclear reactors. The exact nature of this waste is dependent on many factors, as the fuel and its cladding vary with reactor design.

Reprocessing spent fuel generates an acidic liquid HLW which contains the residue of the spent fuel following the removal of nearly all the uranium and plutonium via the Plutonium-URanium EXtraction PUREX process. This residue potentially contains a wide variety of elements derived from four sources, those initially present in the fuel and cladding, those formed during the fission process, a limited number of transuranics formed by neutron capture (e.g. Np, Am and Cm) and chemicals used in the reprocessing. Over the years, many methods have been investigated for the immobilization of commercial HLW, including cements, various glass compositions, glass-ceramics and SYNtheticROCk (SYNROC)-type ceramics, but the first-generation waste form choice was borosilicate glass (Donald *et al.*, 1997).

Compared with the complexity of the commercial wastes, defence wastes are simpler. Generally, defence wastes do not contain the high concentrations of fission products found in commercial wastes, the exception being the calcined naval reactor wastes currently stored at the Idaho National Laboratory (INL), but destined for the Waste Isolation Pilot Plant (WIPP), in New Mexico, USA. They can, however, contain high concentrations of actinides which are not present in commercial wastes. Donald (2007) gave generic compositions for both commercial and defence wastes (see Table 25.2), and although there are very large compositional ranges for the constituents, it does highlight several important differences in addition to those described.

Table 25.2 Generic compositions of typical radioactive wastes streams

Constituent	Commercial waste (mass%)	Defence waste (mass%)
Na_2O	0–39	0–16
Fe_2O_3	2–38	24–35
Cr_2O_3	0–2	0–1
NiO	0–4	0–3
Al_2O_3	0–83	5–9
MgO	0–36	0–1
MoO_3	0–35	0–1
ZrO_2	0–38	0–13
SO_4	0–6	0–1
NO_3	5–25	0
Fission product oxides	3–90	2–10
Actinide oxides	<1	2–23
Other constituents	–	17–27

Source: Donald, (2007).

More specific waste stream compositions are given in Table 25.3 and actual compositions of the solid waste components of various waste streams based on calcine oxide compositions have been reported (Jantzen, 2011) and are shown in Table 25.4.

US wastes derived from the original production of plutonium contain a high concentration of sodium, which arose from the need to neutralize the acidic liquor before it could be stored in the carbon steel tanks built in the early days of the US defence programme at Hanford and the Savannah River Site (SRS). Although neutralization reduced the rate at which the tanks corroded it did not eliminate it entirely, and so these stored wastes contain contaminants from the steel, together with additional iron from the use of ferrous sulphamate in the PUREX process, small concentrations of nickel dissolved from the Ni-plated uranium targets irradiated to produce Pu, and some chromium. Wastes from the other countries involved in nuclear weapons programmes include those generated by the production of Pu and highly enriched uranium (HEU), together with various chemical and pyrochemical processes for reprocessing Pu.

Weapons programmes such as those in the US and the USSR were strictly military and the facilities built to support these programmes were clearly identifiable as were the wastes generated by them. The growing interest in nuclear power for electricity generation meant countries who started nuclear arms programmes only a few years later often had concurrent civil nuclear power and weapons programmes which used the same facilities. For example, in the UK, nuclear reactors at Windscale and Chapelcross were used to generate electricity but at burn-up rates optimized to maximize Pu production. Fuel from these reactors was reprocessed at Windscale along with fuel from other electricity generating reactors. Similarly the UK's gaseous diffusion plant at Capenhurst was used to produce HEU but also lower enrichment grades for civil reactor fuel.

France also had concurrent programmes with the majority of product from its first plutonium separation plant at Marcoule being for military use. In anticipation of a global expansion in plutonium fuelled reactors for civil use, a second plant, at La Hague, was funded by the Commissariat à l'Énergie Atomique's (CEA) civil and military budgets (Schnieder and Marignac, 2008). HLW reprocessing wastes in both France and the UK are being vitrified into steel casks and stored in purpose-built above-ground stores awaiting provision of suitable permanent repositories. In addition to the wastes generated during its production and those arising during the manufacture of warheads, there is also the plutonium which has been declared surplus to requirements following the decision by the US and Russia to reduce their warhead stockpiles. Under the 1993 Non-Proliferation and Export Control Policy (see http://www.fas.org/irp/offdocs/pdd13.htm), the US declared 55 tons of plutonium surplus to national security needs. A similar quantity was

Table 25.3 Specific compositions of a selection of defence and commercial radioactive wastes (gL^{-1})

Component	SRS (USA)*	Hanford (USA)*	Idaho Falls (USA)*	Tokai (Japan)	Sicral 1 (France)	Magnox (UK)	Lanchow (China)
Al	7.7	1.5	4.2	–	32.5	26.0	4.5
Na	5.9	4.1	3.1	44.5	20.5	–	31.0
K	0.3	–	0.9	–	–	–	0.6
Mg	0.2	–	–	–	4.0	30.0	–
Fe	29.7	6.1	–	8.4	16.0	13.0	13.5
Ni	2.8	0.6	–	2.2	1.5	1.4	2.9
Cr	0.3	0.1	–	2.2	1.5	1.6	1.2
Mo	0.2	0.2	–	–	–	10.8	0.7
Zr	0.6	3.4	11.4	–	–	11.8	0.7
Hg	1.8	–	–	–	–	–	–
Cl	0.9	0.1	–	–	–	–	–
SO_4	0.8	0.2	2.6	–	–	–	4.8
NO_3	4.2	2.8	12.5	–	–	11.0 mol	–
Fission products	<3.0	<2.5	<1.0	49.0	24.5	–	2.7
TRU	<0.2	<0.1	<0.1	12.6	3.0	2.0	17.9

*Defence waste.
Source: IAEA (1992).

Table 25.4 Compositions of solids in a selection of defence and commercial radioactive wastes (mass%)

Component	DWPF (Purex HAW) USA*	Hanford 76-68 USA*	West Valley, NY (USA)	Tokai (Japan)	UOX 1 (France)	Magnox (UK)	Myak (Russia)
Al_2O_3	5.99	–	2.39	–	–	19.60	–
CaO	2.46	–	–	–	–	–	–
Cr_2O_3	0.71	1.21	2.92	1.69	3.15	1.6	0.58
CuO	0.17	–	–	–	–	–	–
Fe_2O_3	50.64	29.09	50.30	9.02	18.06	10.0	6.07
HgO	0.33	–	–	–	–	–	–
K_2O	0.16	–	0.44	–	–	–	22.00
MgO	0.56	–	–	–	–	21.6	2.07
MnO	12.70	–	1.34	–	–	–	–
Na_2O	6.33	15.15	6.77	16.46	–	–	27.98
NiO	6.52	0.60	2.01	1.48	2.54	1.20	3.39
PbO	0.68	–	–	–	–	–	–
P_2O_5	0.28	1.52	11.09	0.93	1.76	–	–
SiO_2	1.35	–	–	–	–	–	–
ZnO	0.28	–	–	–	–	–	–
F	–	–	–	–	–	–	–
Cl	0.18	–	0.10	–	–	–	–
SO_4	0.72	–	1.02	–	–	–	–
Fission products	2.71	38.48	3.16	65.01	72.24	44.40	37.91
Actinides	7.24	13.95	18.35	5.41	2.25	1.60	–

Source: Jantzen (2011).
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also declared surplus by Russia. These quantities may be further increased following the 2010 US–Russia strategic arms reduction agreement (see http://news.sky.com/home/world-news/article/15583846).

25.3 Nuclear safety and security

The safe storage and disposal of radioactive waste (RAW) is of paramount importance, with the goal being to convert nuclear wastes into stable solid forms which can be safely stored before permanent disposal in repositories, frequently not yet designed or built, which would serve the dual function of preventing the waste from entering into the biosphere and supporting non-proliferation. Although the primary disposal objective is to protect mankind from the deleterious effects of radiation, most of the elements are toxic or carcinogenic. Important considerations in the storage and ultimate disposal of wastes include not having concentrations of fissile materials that could pose criticality issues, in addition to keeping them resistant to proliferation.

Many proposals for disposing of RAW have been made but most have been excluded for technical, legislative or environmental reasons. Those that remain viable in the foreseeable future are variations on geological disposal with partitioning and transmutation as potential future technologies (e.g., Bowman and Venneri, 1993; King, 2002; González-Romero, 2011).

The option being investigated by most countries is a multi-layer engineered approach in which the processed waste, in a passively safe form, is enclosed within a sealed container and placed underground. Much has been written on the selection of suitable sites (e.g., Ewing, 2004; Murphy, 2004) but the enormous timescales for which performance assessments are required undermine their reliability and Ewing (2004) concludes performance assessments should not be used as the sole justification for the repository's safety.

Continuing safety arguments and public opposition have meant that to date there is only one functioning underground repository for ILW/TRU, the Waste Isolation Pilot Plant (WIPP) near Carlsbad, NM, USA, which accepts defence-related TRU waste, although there are a number of functioning and historical repositories (Rempe, 2007). Several countries have identified sites and are at various stages of developing repositories for the disposal of civil waste, e.g. Finland (expected date 2020), Sweden (2023) and France (2025). The US were developing a repository at Yucca Mountain but, despite spending 23 years and an estimated $13.5 billion, the Administration cut the funding in February 2010 and withdrew the DOE application for a construction permit from the Nuclear Regulatory Commission. As a result of the current impasse in the nuclear waste programme, the government is currently looking at alternatives (Levy, 2010; Schaffer, 2011) and

set up a commission (Blue Ribbon Commission on America's Nuclear Future) in January 2010 to conduct a comprehensive review of policies for managing the back end of the nuclear fuel cycle and recommend a new plan. This report (BRC) was submitted in January 2012 and recommends the building of a temporary storage facility concurrent with work on identifying a site for a permanent geological repository.

Direct injection of liquid waste into deep rock formations has been proposed (Apps and Tsang, 1996; Tsang and Apps, 2005) and the technology was used by the former Soviet Union to dispose of nearly 50 million m^3 at three repositories (Rempe, 2007) in Central European Russia. Disposal of LLW, ILW and HLW at the Krasnoyarsk-26 facility into two aquifers at levels between 180 m and 500 m deep commenced around 1967. An assessment of waste migration by Compton *et al.* (2000) based on official Russian data gathered over many years, concluded that the deep well was functioning as designed and that there is very little likelihood of the waste reaching the surface in sufficient quantity to exceed the drinking water standards. Whilst this is encouraging for the disposal of LLW and some ILW, longer lived isotopes present in defence wastes may make this option problematic.

The possibility of using boreholes as an alternative to underground repositories for the disposal of solid waste has been proposed on several grounds, increased security being one of them. Originally, the proposals were for boreholes to depths comparable to repositories (e.g., 500–700 m), but proposals for ever deeper borehole disposal have been made. One suggestion has been the very deep borehole (Gibb, 1999) in which containers of HLW are placed in a borehole typically 4–6 km deep. The combination of self-heating and geothermal heat would cause either the rock to melt and then encapsulate the waste package (Gibb, 1999) or dissolve the container and contents (Nirex, 2002). As the HLW decays, the rock cools and solidifies with the waste immobilized. The potential for isotopes to leach out of the rock at some very distant timescale has been addressed by the proposal to perform *in-situ* sintering of a mixture of waste and other constituents which would generate a geochemically stable waste form in equilibrium with the host rock (Ojovan *et al.*, 2004). An alternative to the deep borehole is the self-burial technique (Logan, 1999; Ojovan and Gibb, 2005) in which the radiogenic heat generated by the waste melts the rock and the waste package descends.

Although the safety of disposed waste is of great importance in the public's perception of waste disposal, security of the waste, especially weapons grade fissile material, is not significant compared to other concerns such as transport of waste (Solomon *et al.*, 2010). This may be because the quantity of civil fissile material far exceeds the quantity of weapons material. A comparison of the plutonium inventories from the two sources in 2000

suggested nearly 1300 Mt present in the civilian inventory, either in fuel elements or as reprocessed material compared to a military inventory of 255 Mt (Ewing, 2010). This large quantity of civil plutonium forms the basis of the commonly used security standard that was proposed in 1994 by the US National Academy of Science's Committee of International Security and Arms Control (NAS CISAC). The standard requires that 'the surplus weapons useable plutonium should be made as inaccessible and unattractive for weapons use as the much larger and growing quantity of plutonium that exists in the spent nuclear fuel from commercial power reactors (Bunn, 1994).

25.4 Treatment and immobilization

As has been shown in Section 25.2, defence wastes can have an extremely large compositional range, varying from pure metal (i.e., Pu or HEU) through well-defined chemical compounds (e.g., oxides), to the liquid and semi-solid HLW tank wastes found at various separation facilities. Any treatment process must ensure that the ultimate waste form is intrinsically (passively) safe, leach resistant, chemically stable and radiation resistant. The chosen process must also be sufficiently flexible in order to deal with the compositional variability within a waste stream and more significant variations between different varieties of waste stream.

During the Cold War period, the two major protagonists, US and Russia, operated two different philosophies on the value of plutonium which had a significant impact on the inventories of Pu-contaminated wastes and residues requiring disposal (Jardine *et al*., 1999). Whereas US policy was to establish a Pu concentration for the various wastes and residues below which it was considered more economical to produce new metal (economic discard limit, EDL), Russian policy was to recover all Pu above a concentration of 200 ppm for re-use. To implement this philosophy extensive recovery processes were installed at the Pu production facilities of Mayak, Tomsk and Krasnoyarsk.

25.4.1 US tank wastes

In 1996 the first plant built in the US for vitrification of defence-related HLW, the Defence Waste Processing Facility (DWPF) at the Savannah River Laboratory (SRL), commenced operation. Unlike the British and French civil HLW vitrification plants, which operate using Inconel 601-lined induction furnaces, the DWPF operates a Joule-heated ceramic-lined furnace, as will the new facility, the Waste Vitrification Plant (WVP) being built at Hanford. In the DWPF, waste is continually fed into the melter as a wet slurry to minimize dusting of radionuclides along with glass frit and

heated up to 1150°C. The resulting glass is poured into the metal waste canisters at intervals. Borosilicate glasses are used in all of these facilities, but each has its composition tailored to meet the specific requirements of the wastes being processed. Comprehensive tables of glass compositions for both defence and civilian use have been given by Donald (2010) and Jantzen (2011).

Experience gained during the operation of DWPF has led to new glass compositions being developed (Table 25.5) which allow waste loadings to be increased from a nominal 28 mass% to 38 mass% (Marra *et al.*, 2008). These compositions are aimed at the high alumina content wastes to be processed at SRS and Hanford, which are especially problematic due to the refractory nature of alumina which reduces throughput and increases the formation of nepheline ($NaAlSiO_4$) crystals which can be detrimental to the durability of vitreous waste forms by reducing the alumina and silica content of the residual glass. Table 25.5 highlights the variations in composition as glasses are developed for specific waste streams with HAL-17 being developed for Hanford tank sludges containing approximately 53 mass% alumina.

The use of a different furnace technology, the Cold Crucible Induction Melter (CCIM), or skull melter, allows higher vitrification temperatures to be achieved without enhanced corrosion of the refractory liner. Using a small-scale CCIM furnace SIA Radon were able to vitrify SRL Sludge Batch 2 simulated waste at a 50 mass% loading at 1320–1440°C using Frit 320 (Table 25.5) (Stefanovsky *et al.*, 2008). The actual Batch 2 sludge consists primarily of the oxides of Al, Fe, Na and U (Elder *et al.*, 2000).

Table 25.5 Compositions of some borosilicate glasses frits for treating high alumina wastes (mass%)

Melter	Glass	B_2O_3	CaO	Li_2O	Na_2O	K_2O	SiO_2	Reference
DWPF	503	14.0		8.0	4.0		74.0	Marra *et al.* (2008)
DWPF	517	17.0		10.0	3.0		70.0	Marra *et al.* (2008)
DWPF	520	8.0	1.0	10.0	4.0		77.0	Marra *et al.* (2008)
DWPF	521	10.0	1.0	8.0	6.0		75.0	Marra *et al.* (2008)
WVP	HAL-17	31.0	12.0	7.4	4.3	5.0	40.3	Marra *et al.* (2008)
CCIM	Frit 320	8.0		8.0	12.0		72.0	Stefanovsky *et al.* (2008)

25.4.2 Russian wastes

The Pu recovery policy operated by Russia meant that in 1999 there were no substantial inventories of wastes containing >1% Pu (Jardine *et al.*, 1999). Recovery of Pu from wastes led to the generation of waste streams containing <200 ppm Pu which were suitable for cementation and near-surface burial. However, there are some HLW sludges which contain significant concentrations of Pu and various immobilization methods (e.g., vitrification) are being investigated.

25.4.3 Plutonium

Perhaps the easiest material to immobilize, at least technologically, is Pu. It is possible to vitrify Pu in a variety of glass compositions and immobilize it in various ceramic-based hosts. In the latter half of the 1990s, a number of scoping studies were published (e.g., Matzke and van Geel, 1996; Gray and Kan, 1996; Wicks *et al.*, 1996), which outlined a number of potential hosts and demonstrated practically that it was indeed possible to immobilize Pu in a variety of glass and ceramic hosts.

The largest of these studies was performed on behalf of the US DOE which identified 72 possible options (Gray, 1996a) of which five were studied in depth, three involving vitrification and two immobilization in a ceramic host. For the vitrification processes lanthanum borosilicate glass based on the original 'Loeffler' optical glass composition was selected as the host in preference to lead iron phosphate or alkali tin silicate compositions, which had also been considered as potential hosts (Gray, 1996b). Studies of borosilicate glass developed for vitrifying waste arising from reprocessing nuclear fuel elements have shown that this glass can also be used, but suffers from having a low actinide solubility, i.e. <3 mass% Pu, which compares unfavourably with lanthanum borosilicate glass, which can incorporate in excess of 10 mass% PuO_2 (Meaker *et al.*, 1997; Peeler *et al.*, 1997). Two of the vitrification proposals were similar in that the waste plutonium and any scrap material, pre-treated where necessary, would be vitrified in the lanthanum borosilicate glass. However, while one proposal (Gray, 1996b) required a new facility to be built, the second (Gray, 1996c) made use of an existing facility at SRS. In both proposals the glass would be poured into stainless steel HLW canisters, whereas in the third proposal (Gray, 1996d) it would be cast into small metal cans, 20 of which would be carefully positioned within a HLW canister. The size of HLW canisters, 0.6 m diameter × 3 m high and weighing 2,000 kg, mitigates against theft, but additional security was proposed for the first two proposals by spiking the glass with sufficient ^{137}Cs to maintain a γ-radiation field above 1 Gy/hr for 30–60 years. A slightly different approach was proposed for the third option in that the

voidage surrounding the cans would be filled with conventional borosilicate glass containing either ^{137}Cs or HLW. Compositions of a selection of the glasses suggested for immobilizing surplus Pu are given in Table 25.6. Only a few glasses containing radioactive constituents have been prepared, most candidate compositions were made containing non-radioactive rare earth oxides as surrogates for Pu.

Phosphate glasses have also been investigated as they tend to have a higher solubility for actinides than silicate glasses and have been used in Russia as an alternative to borosilicate glass for the immobilization of HLW. Initially they suffered from poor durability and were highly corrosive in the molten state, so they found less favour than borosilicate glasses for which non-active processing technology existed. However, their durability has improved with the development of sodium aluminium phosphate (Minaev *et al*., 2004; Donald *et al*., 2006), iron phosphate (Day *et al*., 1998; Mogus-Milancovic *et al*., 1997) and lead iron phosphate glasses (Sales and Boatner, 1988) and they now have durabilities which match or exceed the standard borosilicate glass. Further information on phosphate-based glasses for immobilizing wastes is given elsewhere (e.g., Donald, 2010; Jantzen, 2011).

The introduction of CCIM technology largely addresses the problem of refractory liner corrosion by containing the melt within a solid skull of glass produced by cooling the outside of the furnace. Corrosion of glass contact refractories used in the vitrification of RAW has been reviewed in, e.g., Bingham *et al*. (2011).

Although the majority of the effort went into investigating vitrification, there was significant effort put into the ceramic option employing a Synroc-based composition which was subsequently selected as the most suitable choice, with disposition to be carried out in a similar manner to that of the vitrification route, i.e., ceramic pellets encapsulated in HLW glass in steel containers. Synroc (synthetic rock) is the generic name for a group of ceramics containing varying proportions of minerals found in nature, including hollandite ($BaAl_2Ti_2O_6$), perovskite ($CaTiO_3$), zirconolite ($CaZrTi_2O_7$) and rutile (TiO_2) (Ringwood *et al*., 1979). Synroc-D was initially developed for defence requirements and consists primarily of perovskite, zirconolite, nepheline ($NaAlSiO_4$) and spinel ($MgAl_2O_4$), together with a continuous intergranular glassy phase, whilst the ceramic developed for immobilizing Pu consisted mainly of zirconolite, the primary phase for incorporating actinide elements. Waste forms with Pu loadings in excess of 10 mass% have been reported to exhibit excellent durability (Jostsons *et al*., 1995). Monolithic waste forms can be produced from a mixture of waste and ceramic precursor powder using conventional ceramic processing techniques, i.e., hot pressing (HP), hot isostatic pressing (HIP) and cold-pressing followed by sintering (CPS). A HP process developed at the Australian Nuclear

Table 25.6 Compositions of some glasses developed for the vitrification of plutonium

Glass	BaO/SrO	La_2O_3	B_2O_3	Al_2O_3	SiO_2	PbO	P_2O_5	Gd_2O_3	Nd_2O_3	PuO_2	Na_2O	Others	Reference
Lan-14	3.5	30.4	4.7	15.0	27.0	11.3						8.2 CeO	Ramsey *et al.* (1994)
Lan-17	3.3	16.2	3.9	22.5	22.5	8.8			20.6			8.3 CeO	Ramsey *et al.* (1994)
LaAlBSi	4.4	23.2	4.3	9.5	26.3	11.1				15.0		6.1 Sm_2O_3 0.2 ZrO_2	Bibler *et al.* (1996)
LaAlBSi	2.5	12.4	11.7	21.5	29.1			8.6	12.8			1.3 ZrO_2	Vienna *et al.* (1996)
LaAlBSi	2.4	8.6	11.4	20.8	28.2			7.5	8.6	11.4			Macfarlane (1998)
LaAlBSi	2.5	20.3	13.0	10.0	20.0			11.7	15.4				Macfarlane (1998)
FEP 1							62.0					38.0 Fe_2O_3	Mogus-Milancovic *et al.* (1997)
FEP 2							85.0					15.0 Fe_2O_3	Mogus-Milancovic *et al.* (1997)
NaAlP1			2.0	19.0			39.0				40.0		Donald *et al.* (2006)
MMW			17.9	6.3	50.4			12.5			8.9	4.1 Li_2O	Harrison *et al.* (2008)
LaBS Frit X	2.5	19.0	13.0	10.0	20.0			13.5	15.0			7.0 HfO_2	Fox *et al.* (2008)

Science and Technology Organisation (ANSTO) utilizes a stainless steel collapsible bellows can into which the mixture is placed. After evacuating and sealing the can, it is cold pressed to approximately two-thirds of its original length before being hot-pressed. Using simulated wastes, ANSTO have demonstrated this process on an industrial scale by successfully producing samples up to 436 mm in diameter.

More recently, both these potential options have been dropped in favour of use of surplus Pu as a mixed oxide (MOX) fuel, in line with the Russian view of Pu as a strategic material rather than a waste, for use in either Pu breeder reactors or light water power reactors (Gong *et al*., 2001; IPFM, 2009). France and Germany started bilateral programmes with Russia in 1992 which demonstrated the feasibility of recycling weapons grade Pu in Russian VVER 1000 and BN 600 reactors (Seyve *et al*., 1999).

The UK government has not declared any weapons grade plutonium to be surplus, but began a public consultation in 2011 (DECC, 2011) into the long-term management of the large stock of UK-owned civilian plutonium, 114.8 te at December 2010 (www.hse.gov). Studies funded by the Nuclear Decommissioning Authority (NDA) into a variety of topics including re-use as MOX fuel, the preferred option, and immobilization will permit decisions to be made on the management of the stocks. Immobilization of civilian stocks declared surplus or unsuitable for re-use in ceramic and vitreous waste forms is being investigated (Harrison *et al*., 2008) and will provide a significant read-over to weapons grade plutonium should a future need arise.

Other alternative disposition options have been suggested, including the use of some surplus Pu to produce ^{99}Mo by irradiation of ^{239}Pu for medical applications (Mushtaq, 2011), but these must generally be viewed as only suitable for dealing with very small quantities.

25.4.4 Pyrochemical wastes

In addition to the issue of treatment of surplus weapons grade materials, increased interest is being shown in the immobilization of special categories of waste arising from the pyrochemical reprocessing of Pu metal for weapons use. These differ from the wastes generated during the reprocessing of spent fuel as they can contain high concentrations of actinides together with substantial quantities of halides, particularly chlorides, as illustrated in Table 25.7, which gives the compositions of simulated salt wastes under investigation at the UK's Atomic Weapons Establishment (AWE).

Wastes containing large quantities of chloride are not amenable to immobilization in borosilicate glass because of the very low solubility of chlorides in this type of glass. Similarly, Synroc-type ceramics cannot be employed either due to low halide solubility.

Table 25.7 Compositions of AWE simulated pyrochemical reprocessing wastes (mass%)

Component	Type I	Type II	Type III	Type IV
HfO_2 (PuO_2 surrogate)		20.7	62.2	11.4
Ga_2O_3		28.0	9.4	10.5
Al_2O_3		9.8	1.7	2.2
Sm_2O_3 (Am_2O_3 surrogate)		4.6	11.7	1.0
MgO		6.3		10.1
FeO		1.5		0.7
Ta_2O_5		1.3		0.7
NiO		1.3		0.7
ZnO				35.7
SiO_2				0.8
B_2O_3				0.8
$CaCl_2$	80.0			
CaF_2		10.4	5.0	8.5
$SmCl_3$ ($PuCl_3$ and $AmCl_3$ surrogate)	20.0			
KCl		16.3	10.0	16.9

Source: Donald *et al.* (2007). British Crown Owned Copyright 2007/AWE.

Two approaches can be taken when dealing with this type of waste. One is to remove the halides prior to immobilization of the non-halide constituents employing, for example, borosilicate or phosphate glass; the second is to accommodate the halides chemically in a suitable host. Halides can be removed by a number of methods, but one disadvantage of this route is that secondary waste is produced which must also be dealt with. One example is reaction of the waste with ammonium dihydrogen phosphate to yield ammonium chloride and water as by-products, together with a phosphate glass (Donze *et al.*, 2000):

$$2NH_4H_2PO_4 + MCl_2 \rightarrow MO.P_2O_5(\text{glass}) + 2NH_4Cl\uparrow + 2H_2O\uparrow$$

Another example is use of lead silicate glass to yield lead chloride as a volatile by-product, the chloride reacting with PbO in the glass, and the resulting oxide dissolving in the glass (Forsberg *et al.*, 1997):

$$3PbO + 2PuCl_3 \rightarrow 3PbCl_2\uparrow + Pu_2O_3$$

Iron phosphate-based glasses have also been suggested for immobilizing chloride wastes, but in the UK AWE's experience, the bulk of the chloride is not incorporated chemically but is evolved during waste form processing, again generating a secondary waste. Calcium aluminosilicate-based glasses have also been suggested (Siwadamrongpong *et al.*, 2004) and have been

shown to be partially effective in incorporating chloride constituents, immobilizing up to 17.5 mol% calcium chloride, for example (Schofield *et al.*, 2008). Unfortunately, regardless of the amount of chloride in the initial batch, up to 30% of the chloride is evolved during the melting process, making this method no more attractive than methods suggested for removing chloride prior to immobilization.

Alternatively, halides can be chemically incorporated into a number of ceramic hosts including chlorapatite, $Ca_5(PO_4)_3Cl$ and spodiosite, Ca_2PO_4Cl, with the actinides being incorporated into the substituted whitlockite-type phase (Donald *et al.*, 2007), which is one of the methods being investigated at AWE. An alternative method involves occluding the halide species into a zeolite and heating above 800°C to form the sodalite mineral phase $Na_8(AlSiO_4)_6Cl_2$ (Lewis *et al.*, 1993; Metcalfe and Donald, 2004). This method has been adopted by the Argonne National Laboratory (ANL) for immobilizing pyrochemical wastes arising from reprocessing of experimental fast breeder reactor fuel, where salt-loaded zeolite is mixed with glass and converted into a monolith by either hot-isostatic pressing or pressureless sintering. The phase assemblage produced by both processes is essentially the same, consisting of about 70% sodalite, 25% binder glass and 5% halite and oxide inclusions (Lewis *et al.*, 2010). A similar method was investigated by AWE for immobilizing weapons-related pyrochemical wastes but was rejected in favour of the phosphate route. In the case of the calcium phosphate immobilization route, waste powder may be reacted with calcium phosphate to yield a mixture of chlorapatite and spodiosite: for example:

$$PuCl_3 + 8Ca_3(PO_4)_2 \rightarrow 2Ca_5(PO_4)_3Cl + 4Ca_2PO_4Cl + Ca_6Pu(PO_4)_6$$

The resulting powder will subsequently be encapsulated in a sodium aluminium phosphate or similar glass to yield a monolithic product.

Fluidized bed steam reforming has also been suggested for treating halide-containing wastes (e.g., Jantzen, 2003). The product from this process is a highly durable waste (Jantzen, 2006) consisting of a number of feldspathoid phases having cage stuctures (e.g., nephelines and sodalite), which contain the halides.

25.4.5 Other defence wastes

In general, radioactive wastes generated by the other nuclear weapons states programmes are less clearly documented.

- France has not declared any plutonium to be excess. HLW from plutonium production has probably been vitrified along with HLW from civilian programmes.

- India has an active nuclear weapons programme (e.g., Chari, 1998). Wastes are probably treated similarly to their commercial nuclear power reactor wastes for which information is available (e.g., Rao, 2001).
- Pakistan also has an active nuclear weapons programme with explosive devices tested in May 1998 (e.g., Kerr and Nikitin, 2011). Information is available on the treatment of radioactive wastes from commercial nuclear power generation programmes (e.g., Hamodi and Iqbal, 2009) and it is likely that defence wastes are treated in a similar manner.
- China has had an active weapons programme for many years and wastes are also likely to be treated in a similar manner to commercial radioactive wastes (e.g., Liangjin *et al.*, 2006).

Waste oils

Waste oils arising from the processing of alpha-emitting materials are problematic as there is limited potential for disposal as solid waste. In the UK, small quantities of some of these wastes containing very low levels of radioactivity have been allowed to be disposed of via incineration at a commercial site (Environment Agency, 2004). However, the incinerator's discharge consent of 80 MBq alpha means this route is incapable of handling all the waste generated. Three methods of treating the wastes have been investigated: biodegradation, acid extraction and electrochemical oxidation. Biodegradation was shown to work, but the amount of LLW generated to dispose of the residual biomass was greater than would have been produced if the oil had been absorbed onto clay granules and cemented (Taylor and Freestone, 2001). The authors consider that with further development, there are potential improvements to the process which would make it a viable process. Using the acid extraction method developed by BNFL Technology Group (now the UK National Nuclear Laboratory) on uranium-contaminated oils, it was shown that repeated washings with sulphuric acid reduced the uranium concentration sufficiently for the oils to be disposed of by controlled exempt release (Environment Agency, 2008). Whilst this process successfully treated uranium-contaminated oils, it was not considered suitable for the corresponding plutonium-contaminated oils because of plutonium's very much higher specific activity and the greater degree of decontamination which is therefore required. More recently, electrochemical oxidation using boron-doped diamond electrodes (Taylor *et al.*, 2009) has been investigated for these wastes and, although it demonstrated potential, it has not been used to date with actinide-contaminated oils.

Fluidized bed steam reforming, in which superheated steam is used as the fluidizing medium, has also been suggested for treating wastes containing organic species (e.g., Williams *et al.*, 2010; Jantzen, 2006). Pyrolysis in the absence of air converts the organics to carbon dioxide.

Highly enriched uranium (HEU)

HEU is another metal of which there is a large stockpile, estimated at 1600 ± 300 tons held globally in 2009 (IPFM, 2009) but not all of which is weapons material, some being present in spent naval and research reactor fuel. The disposal method of choice for unirradiated HEU is blending down to a low enrichment and converting to reactor fuel. It is reported that between 1995 and mid-2009 Russia treated some 367 tons of weapons grade material by this method, and that the US Enrichment Corporation is purchasing 30 tons of blended material from Russia annually (IPFM, 2009).

Tritium

Being a short-lived radionuclide (half-life 12.3 years), tritium gas does not present a disposal problem as it is possible to store it in metal containers until the activity has decayed. Metals have to be carefully selected as they need to be able to cope with the effects of embrittlement by both the tritium and the ^{3}He decay product in addition to a doubling of pressure caused by the formation of ^{3}He. It is also possible to immobilize the gas as a metal hydride and this can have advantages over the gaseous form for long-term storage (Holtslander and Yaraskavich, 1981; IAEA, 2004). Titanium and zirconium are two metals whose hydrides are suitable because they have low dissolution pressures and are reasonably stable to air and water.

25.5 Waste form properties

Many properties need to be considered in waste form development. Mechanical properties are important from the point of view of material integrity in a storage or disposal environment. Of particular importance is the long-term chemical durability of the waste form as this will influence the release of toxic elements due to leaching under disposal conditions. Chemical durability can also change over very long timescales depending on the radiation stability of the waste form, particularly as this may adversely affect the dissolution rates through, for example, the generation of corrosive radiolysis products in the leaching solution.

Durability studies performed using a modified MCC-1 procedure on AWE monolithic material which had undergone accelerated ageing trials through the substitution of ^{238}Pu for ^{239}Pu demonstrate this adverse effect. ^{238}Pu-doped material which had been aged for 1820 days (total α radiation fluence of $4 \times 10^{18}\,g^{-1}$) was leached in water at 40°C for 28 days and compared with ^{239}Pu-doped material (unaged) and the results are summarized in Table 25.8. It can be seen that the release of elements from the aged ^{238}Pu

Table 25.8 Normalized elemental mass loss from aged and un-aged waste simulant

	Normalized elemental mass loss (gm^{-2})				
	Ca	P	Cl	Pu	Am
^{239}Pu unaged	1.6×10^{-3}	2.3×10^{-3}	2.7×10^{-3}	1.2×10^{-5}	2.4×10^{-7}
^{238}Pu unaged	1.6×10^{-4}	5.4×10^{-5}	2.0×10^{-2}	1.6×10^{-5}	$<8.0 \times 10^{-7}$
^{238}Pu aged	1.9×10^{-2}	1.7×10^{-2}	8.8×10^{-2}	1.4×10^{-3}	4.7×10^{-4}

Source: Metcalfe *et al.* (2009).

samples is considerably greater than that from the unaged 238 or 239 samples.

25.6 Future trends

Although current radioactive waste forms used for immobilizing commercial wastes are reasonably well characterized, this is not the case for some of the more recent types of defence wastes, particularly those generated during the pyrochemical reprocessing of Pu metal. In the case of ultimate waste form disposal, longer term predictive capabilities out to geological timescales are required coupled with a need to bridge the gap between computer models and experimental data. Advanced methods for characterizing wasteforms, in order to understand long-term behaviour better, are also needed. Alternative methods of treatment are also under investigation, including partitioning of wastes into short- and long-lived species, with immobilization in borosilicate glass as an option for the short-lived isotopes and Synroc-type ceramics for the long-lived species. Transmutation has also been considered as a means of treating long-lived isotopes by converting them into shorter lived or more stable species. Although not currently a cost-effective method of treatment, this method may offer scope as a future technology for dealing with particularly difficult species.

Dealing with the wastes generated by the pyrochemical reprocessing of weapons Pu is important not just from a defence viewpoint, but is also likely to be increasingly important in the future when the newer Gen IV power reactors are built, as these will be closed fuel cycle systems requiring reprocessing by pyrochemical methods of spent fuel.

25.7 Sources of further information

There are a number of reviews and related articles outlining the various methods of dealing with defence wastes. See, for example, Donald (2010),

chaps 5 and 9, NNSAM (2002), IPFM (2009), the report by the Committee on International Security and Arms Control (1994), and the reviews by Hench *et al*. (1984), Wicks (1985) and Marples (1996). There are many recent reviews on the immobilization of radioactive wastes in general, including chemical durability and radiation stability; see, for example, Donald (2010); Weber *et al*. (2009); Caurant *et al*. (2009); Ojovan and Batyukhnova (2007); Jantzen (2011); Ojovan and Lee (2005, 2007).

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26

Modeling and strategy approaches for assessing radionuclide contamination from underground testing of nuclear weapons in Nevada, USA

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Abstract: This chapter outlines the hydrogeological setting of the Nevada National Security Site (NNSS) and the expected pathways of groundwater flow and radionuclide transport. It describes the evolving strategy developed cooperatively between the National Nuclear Security Administration Nevada Site Office (NNSA/NSO) and the Nevada Division of Environment Protection (NDEP) to assess groundwater contamination from underground testing of nuclear weapons and to protect the health and safety of the public. The modeling challenges and progress in the Underground Test Area Project (UGTA) are also discussed.

Key words: radionuclide contamination, groundwater, flow and transport model, regulatory strategy.

26.1 Introduction

The Underground Test Area Project (UGTA) of the US Department of Energy (DOE), National Nuclear Security Administration Nevada Site Office (NNSA/NSO) is implementing remediation strategies for protecting the health and safety of the public and the environment from radioactive contamination of groundwater produced during past underground testing of nuclear weapons at the Nevada National Security Site (NNSS; formerly called the Nevada Test Site). The NNSS was chosen as the continental site for testing nuclear weapons in 1950 because of the sparse population in the arid southwest region of the United States, the availability of nearby facilities for operational support, and to reduce the cost and logistical difficulties of testing in the western Pacific (US Department of Energy (DOE), 2000a). The first atmospheric tests were conducted in 1951 and the NNSS subsequently became the primary site for testing nuclear weapons. Following the Limited Test Ban Treaty of 1963, atmospheric testing ceased, and nearly 90 percent of the underground weapons tests by the United States were detonated at the NNSS (USDOE, 2000a). Congress imposed a moratorium on

testing of nuclear weapons, and in September of 1992, underground testing ceased.

The NNSS continues to be used for national defense activities and is a major remediation site for the DOE Environmental Management mission of cleanup of the environmental legacy from nuclear weapons and nuclear energy research. The Environmental Restoration Project was established in 1989 for evaluating and remediating contaminated sites on the NNSS and other areas of the state of Nevada. The UGTA under the Environmental Restoration Project is tasked with assessing contaminated groundwater from underground testing. The NNSA/NSO also operates and maintains two facilities located in alluvial basins of the NNSS that dispose of low-level radioactive waste (RAW) and mixed low-level radioactive waste. The RAW is from cleanup activities on the NNSS and from cleanup activities at multiple remediation sites across the DOE complex (nationwide). The RAW is buried in shallow trenches, pits, subsidence craters created by underground testing of nuclear weapons and large-diameter boreholes (greater confinement boreholes) (Shott *et al*., 1998, 2000; Crowe *et al*., 2002, 2005; USDOE, 2005).

The UGTA is evaluating 907 underground nuclear detonations that were conducted at the NNSS; all underground tests are listed in a compendium of weapons tests conducted by the United States from July 1945 through September 1992 (USDOE, 2000b). The NNSS tests were conducted above, near and below the groundwater table in alluvial basins, in volcanic highlands, in shafts and tunnels of zeolitized volcanic rocks, and in tunnels mined in granitic rock.

The phenomenology of underground nuclear explosions is summarized in Borg *et al*. (1976), US Congress Office of Technology Assessment (1989), and the International Atomic Energy Agency (IAEA, 1998). An underground test produces a spherical cavity from combined vaporization, melting and shock compression of the host rock. As the detonation pressure subsides, the rocks above the cavity typically collapse (timeframe of seconds to days after the test) and the cavity is filled with rubble consisting of collapsed rock, and solidified rock melt (melt glass). The collapse void can propagate upward variable distances forming a chimney that may or may not extend to the surface forming a subsidence crater. The temperature and pressure history of an explosion and response of the surrounding host rock control the distribution of radionuclides around the test. Radionuclides produced underground include tritium, fission products, actinides and activation products. Refractory radionuclides (higher boiling points) are trapped primarily in the melt glass, and in cavity rubble and compressed rock around the cavity (up to 1.5 cavity radii from the test point); volatile species circulate outward and condense in cracks and void spaces for distances of 1–3 cavity radii from the test point (Tompson *et al*., 1999; Tompson, 2008; Pawloski *et al*., 2008).

The radionuclides deposited underground from detonation of a nuclear device are referred to as the radiological source term; the portion of the inventory that is migrating in groundwater is the hydrological source term, a subset of the radiological source term. Underground testing on the NNSS deposited an estimated 132 million curies of radioactivity below ground, decay corrected to 1992 (the radiological source of Bowen *et al.*, 2001). Unclassified estimates of this radiological inventory are apportioned among 43 radionuclides and these radionuclides define the source term used in the modeling studies.

Important features of the NNSS with respect to radionuclide contamination of groundwater are the considerable depth from the surface to groundwater throughout most of the site and the absence of natural springs or surface areas of groundwater discharge on the NNSS which would allow radioactive contaminants to be released in the environment. Accordingly, there are no immediate hazards to workers or the public from exposure to contaminated groundwater. The challenges facing the UGTA are to understand the physical and chemical processes of migration of radionuclides within and adjacent to the NNSS, to forecast migration of radionuclides over 1,000 years, and to support regulatory decisions to protect the public. The approach used to address these challenges is a combination of data collection and development of numerical models of groundwater flow and radionuclide transport, model evaluation to test and build confidence in model results sufficient to design a long-term monitoring network, and identification of institutional control policies to restrict public access to contaminated groundwater.

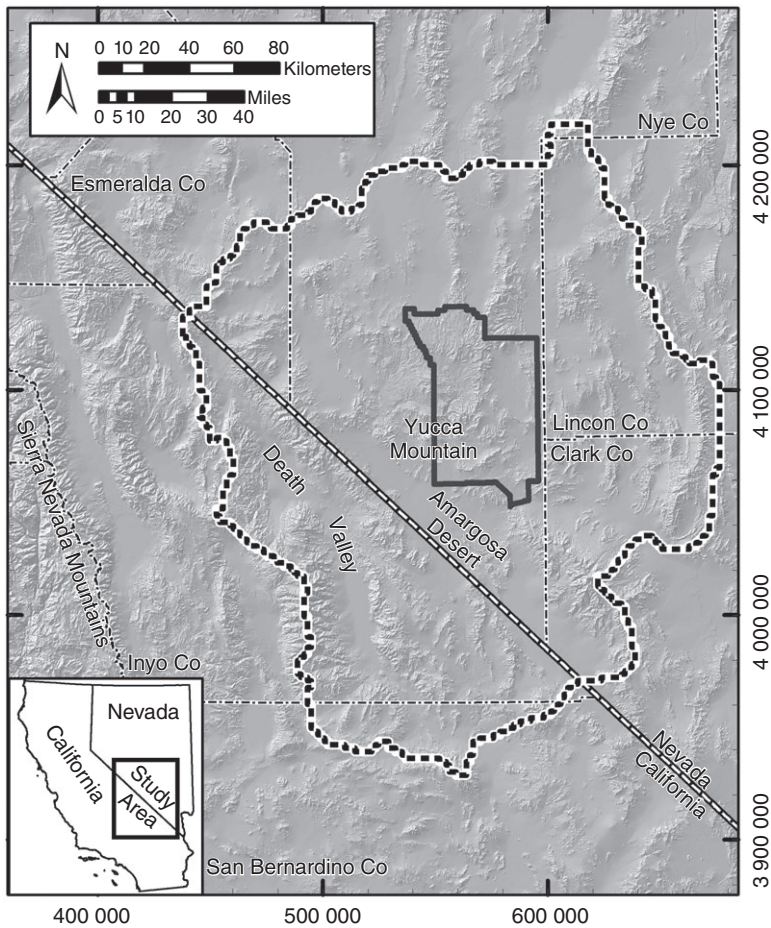
The goals for this chapter are:

- to describe the hydrogeological setting of the NNSS and the expected pathways of groundwater flow and radionuclide transport,
- to describe the evolving strategy developed cooperatively between the NNSA/NSO and the Nevada Division of Environment Protection (NDEP) to assess groundwater contamination from underground testing of nuclear weapons and to protect the health and safety of the public,
- to describe the modeling challenges and progress in UGTA.

26.2 Hydrogeological setting of the Nevada National Security Site (NNSS)

The NNSS is located in the Great Basin portion of the basin-range physiographic province of the southwestern United States (Hunt, 1967; Stewart, 1980), approximately 150 km east of the Sierra Nevada mountain range containing the highest point in the contiguous United States (Mount Whitney, 4421 m) and about 40 km northeast of Death Valley, the lowest

point in North America (86 meters below sea level) (see Fig. 26.1). There are multiple definitions of the Great Basin based on hydrographic, physiographic, and floristic criteria (Grayson, 1993), but the most useful definition for this chapter is the hydrographic definition. The Great Basin is an area centered about the state of Nevada, and including parts of the states of California, Utah, Oregon, and Idaho of the western United States that are internally drained. Precipitation in the Great Basin has no ocean outlet



26.1 Shaded relief map of Southern Nevada, and adjacent areas of California in the southwestern United States. The solid line denotes the boundary of the Nevada National Security Site (NNSS; formerly the Nevada Test Site). The dashed line is the boundary of the Death Valley regional flow system (DVRFS) after Belcher and Sweetkind (2010). Death Valley and the Amargosa Desert are major discharge areas for the DVRFS.

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and surface drainage flows into ephemeral streams that empty into saline lakes or dissipate through combined evaporation, transpiration and/or infiltration. Groundwater flow is an important component of the regional water budget and the NNSS is located in the central part of the Death Valley regional flow system (DVRFS; Winograd and Thordarson, 1975; D'Agnese *et al*., 1997; Belcher *et al*., 2004), a large internally drained area of Nevada (Fig. 26.1). Recharge in the DVRFS occurs primarily at higher elevation mountain ranges in the north, east and southern parts of the flow system. Discharge areas are distributed in the lower elevations of the Amargosa Desert and ultimately Death Valley (Fig. 26.1). The climate of the region is arid and is controlled largely by the rain shadow of the Sierra Nevada mountain range to the west with local variations controlled by elevation.

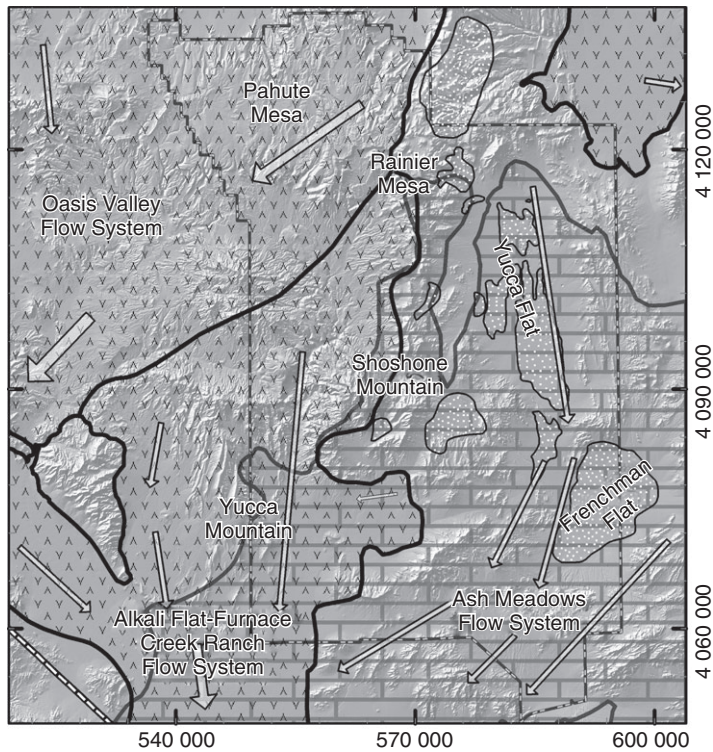
The hydrology of the NNSS is controlled primarily by three hydrologic and geological features. The first is the underflow of groundwater in the DVRFS and the location of areas of significant local recharge in the NNSS at the higher elevation mountain ranges and mesas of the site. The second feature is the physical properties and spatial distribution of diverse assemblages of rock lithologies that form the aquifers and aquitards for the groundwater flow system. These rocks comprise three major lithologically and temporally distinct groups including Paleozoic carbonates and clastic sedimentary rocks, Miocene volcanic rocks erupted from multiple coalesced caldera centers, and thick alluvium deposited in fault-controlled basins. The third feature is the location and nature of major structural and tectonic features, including regional thrust belts formed in late Paleozoic and Mesozoic time, major structures associated with caldera collapse and resurgence, and Miocene and younger extensional and strike-slip faults that formed the alluvial basins of the eastern and southern areas of the NNSS. This combination of features control the volume, velocity and direction of groundwater flow and resulting transport of testing-introduced radionuclides.

26.2.1 Eastern carbonate aquifer

The eastern and southeastern region of the NNSS is characterized by groundwater flow predominantly within a regional carbonate aquifer (Fig. 26.2) that is part of an extremely thick, sedimentary sequence of Neoproterozoic to mid-Paleozoic (Devonian) marine carbonate and clastic rocks deposited at the western edge of a stable continental margin of the North American craton (Sweetkind *et al*., 2010). The carbonate aquifer is the primary aquifer throughout much of the DVRFS (Mifflin and Hess, 1979; Belcher *et al*., 2004). This assemblage of mostly mid-Paleozoic rocks was deformed by Late Paleozoic and Mesozoic regional thrust faults which redistributed the assemblages of carbonate and clastic sediments (Cole,

1997; Cole and Cashman, 1999). These thrust-disrupted rock assemblages were intruded by small granitic stocks during the Late Cretaceous. The thrust faults are exposed at the surface primarily in and west of Yucca Flat; these rocks mark the approximate western boundary of carbonate-dominated groundwater flow (Fenelon *et al.*, 2010).

Thick fanglomerate, alluvial deposits and the distal facies of eastward thinning volcanic rocks uncomformably overlie the Paleozoic sedimentary rocks in the extensional basins of Yucca and Frenchman Flat (Fig. 26.2), two of the major sites of underground testing.



26.2 Major hydrological flow systems of the NNSS modified from Fenelon *et al.* (2010). Regional groundwater flow in the eastern NNSS is primarily through the regional carbonate aquifer (brick pattern) with local contributions from isolated alluvial and volcanic aquifers (v-stiple pattern). The discharge areas for this flow system are Ash Meadows and Alkali Flats/Death Valley. Groundwater flow in the western NNSS is primarily in volcanic aquifers (v patterns) with discharge areas in Oasis Valley and Alkali Flats/Death Valley. These two flow systems control the migration of radionuclides produced during underground testing of nuclear weapons on the NNSS.

26.2.2 Western Volcanic Highland

The geology of the western region of the NNSS consists primarily of thick sequences of ash-flow tuff, lava, and volcaniclastic rocks deposited during episodic volcanic cycles associated with the formation of as many as six coalesced caldera centers from 15 million to 9 million years ago (the Southwest Nevada Volcanic Field; Byers *et al*., 1976; Sawyer *et al*., 1994). These caldera centers are localized in the north-northeast trending Amargosa Desert rift zone (Wright, 1989; Carr, 1990; Fridrich, 1998), a major north-northeast trending structural trough identifiable using gravity and seismic reflection data (Healey *et al*., 1980; Brocher *et al*., 1998). The thick volcanic section in the Amargosa Desert rift zone locally replaces the carbonate aquifer as the primary pathway for regional groundwater flow. The carbonate aquifer is either missing, too deep in the stratigraphic section and/or impermeable from contact metamorphism associated with caldera plutonism to transmit significant quantities of groundwater. Regional groundwater flow in the western volcanic sequence is topographically controlled and driven by the increased recharge at higher elevations, primarily from eastern Pahute Mesa (Blankennagel and Weir, 1973; Laczniak *et al*., 1996; SNJV, 2009a; Fenelon *et al*., 2010).

The Miocene volcanic rocks form high elevation plateaus of welded and nonwelded ash flow sheets concentrically flanking their source calderas. This plateau topography remains preserved where basin-range deformation has locally faulted but has not significantly extended and disrupted the mesas (Pahute and Rainier mesas and Yucca Mountain) (Fig. 26.2). This layered sequence of outer caldera ash-flow sheets is replaced locally by thick sequences of densely welded ash-flow tuff and intrusive rocks within caldera depressions. This pattern of extra- and intra-caldera rock sequences is complicated in the northern part of the Amargosa Desert rift zone by multiple stages of caldera formation. Younger calderas disrupt and bury the structure and volcanic rock assemblages of older calderas.

Spatial changes in lithology and thickness of the volcanic rocks of the western volcanic highland are significant (laterally and vertically heterogeneous), and they are locally affected by secondary alteration (zeolitization), burial diagenesis and/or hydrothermal activity. These lithologic and alteration features strongly affect the hydrologic properties of the rocks (tend to reduce conductivity) and form complex inter-layered aquifers and confining units locally offset or truncated by caldera structures and/or extensional faults. Groundwater flow can be rapid (tens of meters per year) within zones of higher density cooling joints within welded tuff and rhyolite lavas, both augmented by flow along faults; groundwater flow is much slower (one meter per year or less) through altered volcanic rocks and/or zones of matrix-dominated permeability.

26.3 Underground testing and groundwater flow and transport in corrective action units

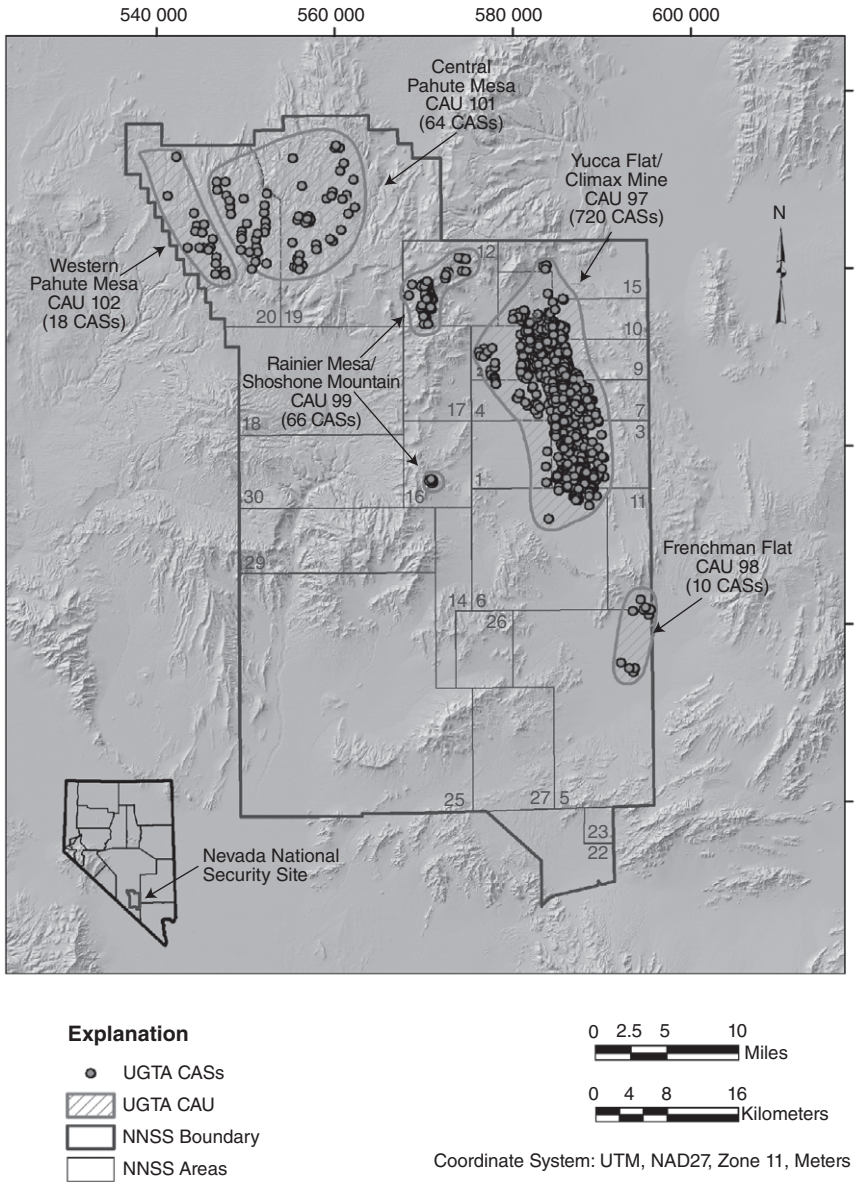
The following sections briefly describe the hydrogeological setting by correction action units (CAUs) of geographical areas used for underground testing on the NNSS (Fig. 26.3).

26.3.1 Frenchman Flat CAU

Ten underground detonations were conducted in Frenchman Flat (Figs 26.3 and 26.4), a strike-slip pull-apart basin formed at the northeastern termination of the Rock Valley fault (Bechtel, Nevada (BN), 2005). Seven tests were detonated in the northern part of the basin in the lower part of the unsaturated zone in alluvium and distal facies of the volcanic rocks originating from eruptive centers in the volcanic highland to the northwest. Three tests were conducted in alluvium in central Frenchman Flat; two of the tests are in the unsaturated zone and one test was detonated below the water table (NNES, 2010a). Local directions of groundwater flow in the Frenchman Flat basin are difficult to establish because of low hydrologic gradients in the basin. Flow is inferred to be predominantly to the southeast driven by higher groundwater levels northwest of Frenchman Flat across the northeast trending, right-slip Cane Spring fault (NNES, 2010a) (Fig. 26.4). Groundwater velocities are very low (1 meter per year or less) down gradient of nuclear tests conducted in alluvium (high porosity alluvial aquifer) in central and northern Frenchman Flat, but may be higher down gradient of two tests where flow is in fractured volcanic aquifers (welded tuff and basalt lava; SNJV, 2006; NNES, 2010a). Gradients in the alluvial and volcanic aquifers are downward but flow from these sections into the underlying carbonate aquifer is limited across a basal confining unit of zeolitized volcanic rocks.

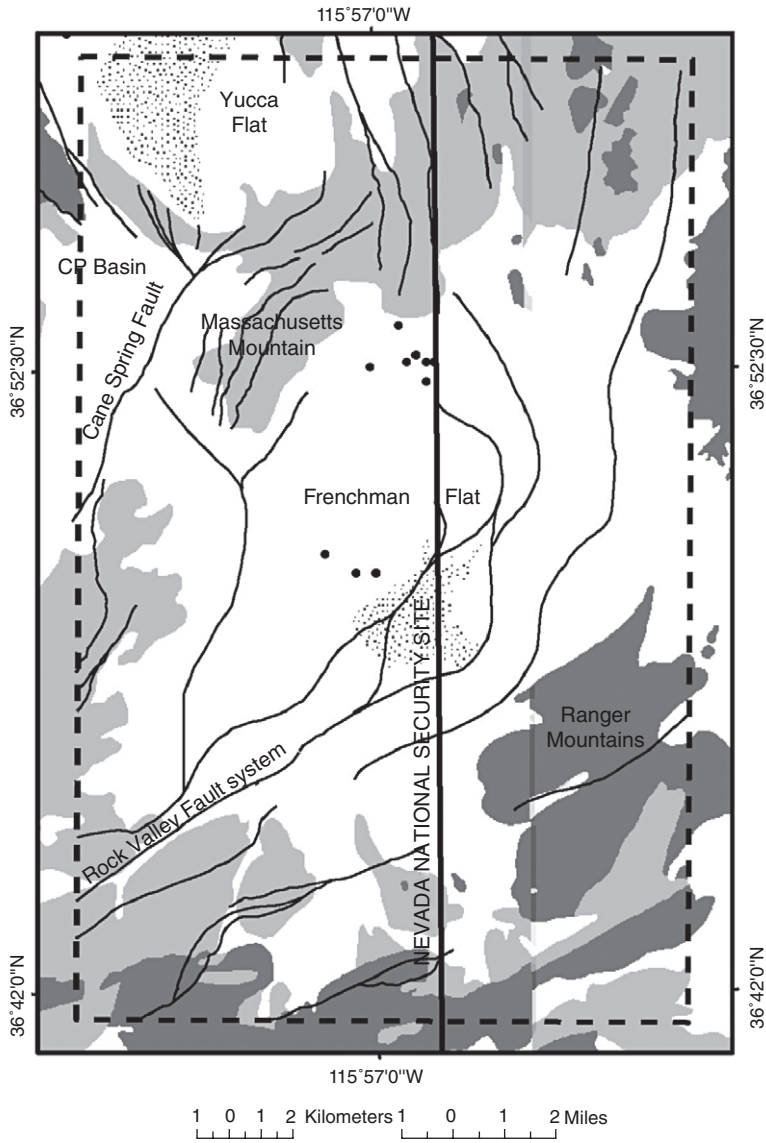
26.3.2 Yucca Flat CAU

A total of 747 underground detonations were conducted in Yucca Flat (USDOE, 2000b), an extensional basin located north of Frenchman Flat in the eastern NNSS (Figs 26.3 and 26.5). Some 664 were in alluvium and volcanic rocks in the unsaturated zone; 76 were in saturated alluvium and volcanic rocks; four in carbonate rocks with two of the detonations in the unsaturated zone and two below the water table; three detonations were in granitic rock in a small Cretaceous stock at the north end of the Yucca Flat basin (Pohlman *et al.*, 2007). The radiological source term for detonations in the unsaturated zone remains in the unsaturated zone with two exceptions. Detonations near the water table may directly inject radionuclides



Source: Appendix VI, Revision No. 4 of the FFACO, 1996 (as amended April 2011)

26.3 Shaded relief map of the Nevada National Security Site showing the location of sites of underground testing of nuclear weapons. The 907 underground detonations are identified as corrective action sites, a subset of the number of underground detonations. Clusters of corrective action sites are grouped into corrective action units (CAUs) and the hydrology and geology of the four major CAUs are described in this chapter.



26.4 Generalized geologic map of the Frenchman Flat basin of the southeast Nevada National Security Site showing the domain area for numerical models of groundwater flow and radionuclide transport at sites of underground testing. Stiple = Quaternary playa deposits; white = Quaternary/Tertiary alluvium; light gray = Miocene volcanic rocks; cross-hatch = Quaternary/Pliocene basaltic rocks; dark gray = Precambrian and Paleozoic sedimentary rocks. Dashed line is the Frenchman Flat hydrostratigraphic framework model boundary. Solid line is the Nevada National Security Site boundary. Dots show the location of ten underground nuclear detonations in the Frenchman Flat corrective action unit.

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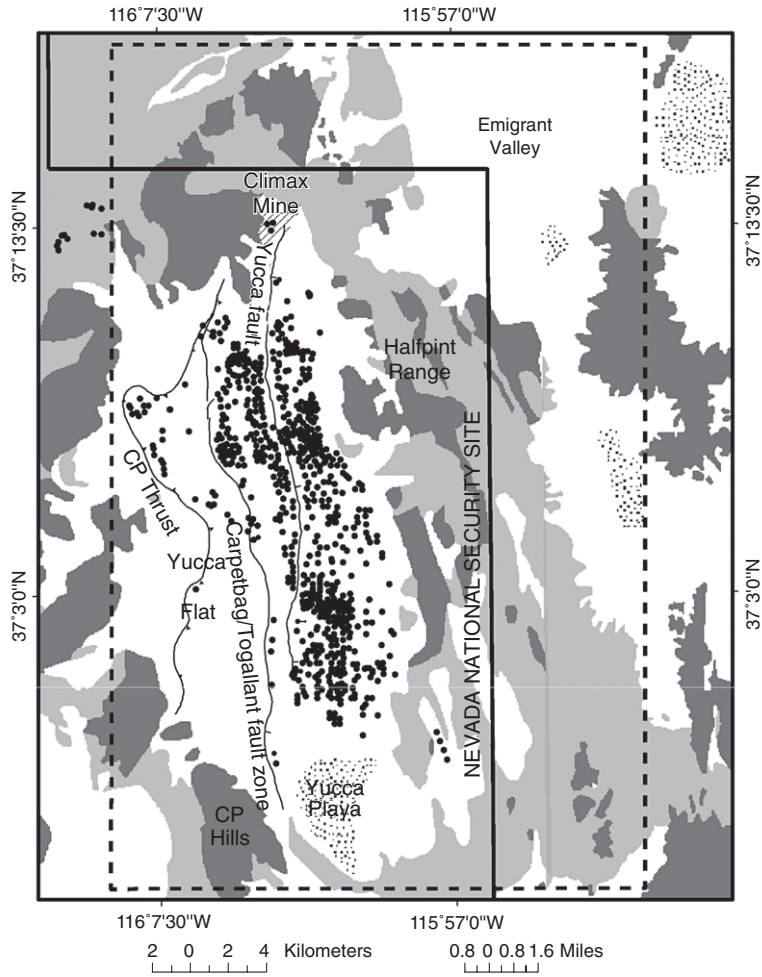

26.5 Generalized geologic map of the Yucca Flat-Climax Mine (YF-CM) corrective action units showing the domain area for numerical models of groundwater flow and radionuclide transport at sites of underground testing. Stipple = Quaternary playa deposits; white = Quaternary/Tertiary alluvium; light gray = Miocene volcanic rocks; diagonal line = Mesozoic granitic rocks; dark gray = Precambrian and Paleozoic sedimentary rocks. Dashed line is the YF-CM hydrostratigraphic framework model boundary. Solid line is the Nevada National Security Site boundary. Dots show the location of 747 underground detonations in the Yucca Flat-Climax Mine corrective action units.

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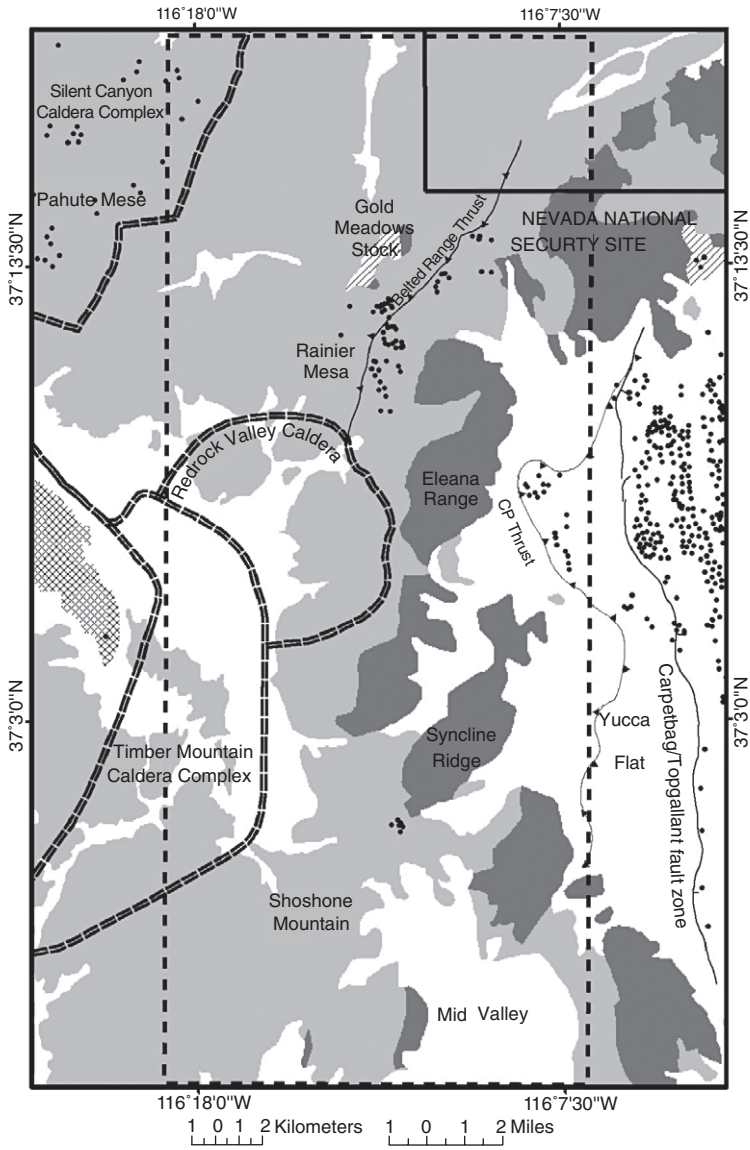
into the water table. Underground detonations that created surface subsidence craters can accumulate surface runoff in the craters. Enhanced infiltration in the crater bottoms moves downward in the collapse chimneys through the test cavity of underground detonations, and may transport radioactive contaminants to the saturated zone. Similarly, underground tests in the unsaturated and saturated zone of subsurface volcanic rocks may directly inject radionuclides into the underlying carbonate aquifer or radionuclides may move downward along local faults and fractures. Local flow of groundwater may transport radionuclides along faults driven by transient pressure gradients created by pressurization of low permeability zeolitized volcanic rocks during underground testing. The phenomenology of underground tests detonated in carbonate rocks is significantly different from tests conducted in other rocks types (Carle *et al*., 2008; SNJV, 2008). The thermal decomposition of carbonate rocks releases large quantities of CO_2 gas that contributes to pressure and density-driven flow. Additionally, radionuclides released in saturated carbonate rocks may be transported directly in the regional groundwater flow system.

Groundwater flow along the length of the Yucca Flat basin is limited by restricted regional underflow from a combination of confining units bounding the basin on the north (granitic confining unit), on the northeast (lower clastic confining unit) and on the west (upper clastic confining unit) (Laczniak *et al*., 1996; Bechtel Nevada, 2006). Recharge in the basin interior is low from the arid climate and downward drainage to the LCA is additionally restricted by the presence of a thick and continuous tuff confining unit at the base of the volcanic section above the carbonate aquifer. Directions of groundwater flow in the alluvial and volcanic aquifers in the Yucca Flat basin are variable and these flow systems are incompletely coupled to the carbonate aquifer (Fenelon *et al*., 2010)

26.3.3 Rainier Mesa/Shoshone Mountain CAU

A total of 68 underground detonations were conducted in tunnels constructed in unsaturated zeolitized volcanic rocks of Rainier Mesa and Shoshone Mountain (tunnel beds); all were located well above the regional groundwater table. Two detonations were in vertical shafts near the water table. The Rainier Mesa and Shoshone Mountain sites form plateau highlands that demarcate the approximate eastern edge of the thick accumulations of volcanic rocks formed within the Amargosa Desert rift zone (Figs 26.3 and 26.6). The migration pathway of radionuclides released during testing beneath Rainier Mesa is generally downward through the unsaturated zone, complicated by local zones of perched water; travel time to the regional water table may be substantial. Additionally, there are local losses of radionuclides from drainage into tunnels constructed to host the



26.6 Generalized geologic map of the Rainier Mesa-Shoshone Mountain (RM-SM) corrective action unit showing the domain area for numerical models of groundwater flow and radionuclide transport at sites of underground testing. White = Quaternary/Tertiary alluvium; light gray = Miocene volcanic rocks; cross-hatch = Quaternary/Pliocene basaltic rocks; diagonal line = Mesozoic granitic rocks; dark gray = Precambrian and Paleozoic sedimentary rocks. Dashed line is the RM-SM hydrostratigraphic framework model boundary. Solid line is Nevada National Security Site boundary. Double-dash line is caldera structural margin (buried). Dots show the location of 68 underground detonations in the Rainier Mesa-Shoshone Mountain corrective action unit (as well as those in western Yucca Flat and eastern Pahute Mesa).

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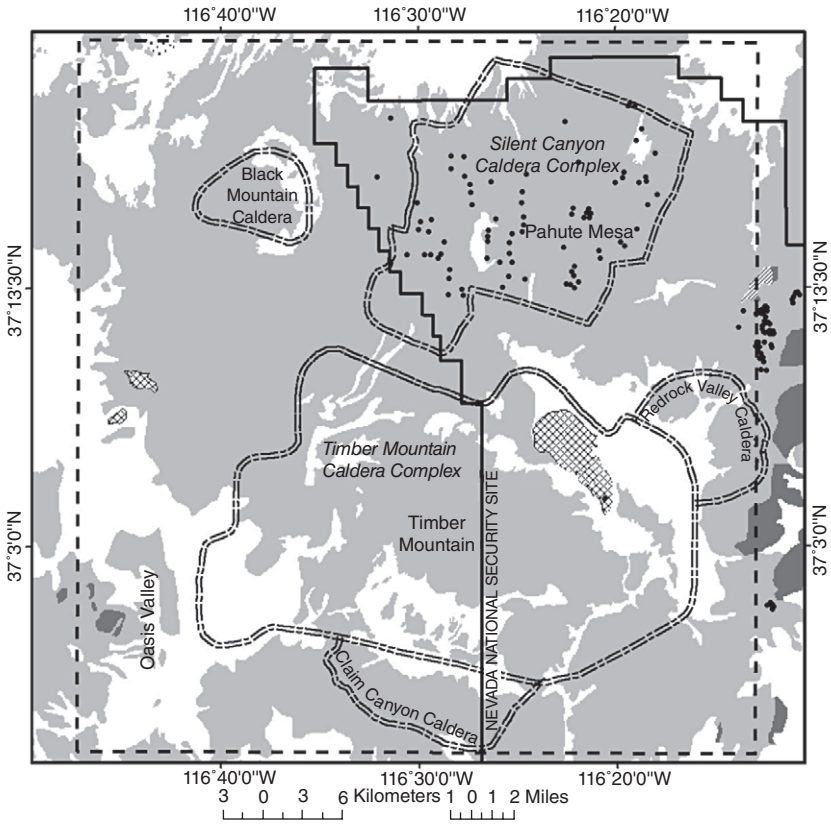
underground tests and from discharges from the tunnels into unlined drainage ponds.

Groundwater beneath Rainier Mesa and Shoshone Mountain is recharged primarily by infiltration through the thick unsaturated zone beneath volcanic highlands; downward infiltration through the zeolitized tunnel beds is locally aided by flow in zones of discontinuous fractures forming local perched water (Russell *et al*., 1987). The amount of underflow beneath the mesa from regional groundwater flow of the DVRFS is poorly constrained and the recharge mound beneath the mesa highlands may not be well connected to the regional groundwater flow system because of local juxtaposition of clastic sedimentary confining units associated with thrust faults at the eastern edge of Rainier Mesa (Fenelon *et al*., 2008).

26.3.4 Pahute Mesa CAU

There were a total of 82 detonations on Pahute Mesa; 64 were located in the central or eastern part of the mesa and 18 were located in western Pahute Mesa. The detonations were in a variety of rock types ranging from confining units of zeolitized volcanic rocks to fractured lava flow and welded tuff aquifers. Pahute Mesa is a large plateau highland formed from the successive eruption of overlapping ash-flow sheets and local silicic lavas from at least six large collapse calderas (Figs 26.3 and 26.7). Three of the calderas are partly to completely covered by volcanic rocks from younger caldera cycles. The down gradient connectivity of the different rock types at the detonation depth strongly affects the local release and rate of groundwater transport of radionuclides from the underground tests. Transport of radionuclides is locally aided by multiple sets of north-northeast trending basin-range faults and may be aided or impeded by offsets of rock units along the basin-range faults or across volcanic structure (ring-fracture zones bounding zones of caldera collapse).

Groundwater flow beneath Pahute Mesa is controlled by underflow from the DVRFS and local recharge at the higher elevations of the eastern mesa areas. Flow is predominantly from higher topography on the northeast to lower topography on the southwest. Local diversions in directions of groundwater flow occur near basin-range faults (Blankennagel and Weir, 1976) and from juxtaposition of confining units and aquifer units across the basin-range faults and caldera structure (SNJV, 2009a). The resurgent dome of Timber Mountain south of Pahute Mesa (Fig. 26.7) diverts groundwater flow to the east or west from a combination of reduced permeability of volcanic rocks associated with intrusion of a granitic body beneath the resurgent dome and/or local recharge at higher elevations of Timber Mountain. Groundwater flow west of Timber Mountain follows the western ring-fracture zone of the Timber Mountain caldera and local basin-range faults,



26.7 Generalized geologic map of the Pahute Mesa corrective action units showing the domain area for numerical models of groundwater flow and radionuclide transport at sites of underground testing. Stiple = Quaternary playa deposits; white = Quaternary/Tertiary alluvium; light gray = Miocene volcanic rocks; cross-hatch = Quaternary/Pliocene basaltic rocks; diagonal line = Mesozoic granitic rocks; dark gray = Precambrian and Paleozoic sedimentary rocks. Dashed line is the PM-OV hydrostratigraphic framework model boundary. Solid line is the Nevada National Security Site boundary. Double-dashed line is the caldera structural margins. Dots show the location of 82 underground detonations in the Pahute Mesa corrective action units (as well as those in the Rainier Mesa CAU).

moving south and southwest to discharge areas of Oasis Valley (USDOE, 1997; Grauch *et al*., 1999). A smaller component of flow may be diverted around the eastern flanks of Timber Mountain, following the Fortymile Wash drainage beneath eastern Jackass Flats and reaching discharge areas of the Armagosa Valley (Fig. 26.2; SNJV, 2009a). A component of flow in western Pahute Mesa may be in carbonate rocks in the vicinity of the Black Mountain caldera west of and outside the Amargosa Desert rift zone. Here

groundwater flow remains west of the Purse fault, a probable hydrologic barrier, but merges with the recharge water from eastern Pahute Mesa near the juncture of the multiple coalesced calderas on the southwest edge of Pahute Mesa (Blankennagel and Weir, 1976; SNJV, 2009a).

26.3.5 Radionuclide transport pathways off of the Nevada National Security Site

Regional groundwater flow in the eastern NNSS is southward through the carbonate aquifer beneath the basins and testing areas of Yucca and Frenchman Flats (Fig. 26.2). Groundwater flow directions change to the southwest in southern Frenchman Flat influenced by increased underflow from east of the NNSS, and following en echelon faults of the southwest trending, right slip Rock Valley fault system (USDOE, 1997; O'Leary, 2000; Belcher *et al*., 2004). The eastern carbonate flow system of the NNSS drains either into the Alkali Flat-Furnace Creek Ranch or Ash Meadows discharge areas of the southern Amargosa Valley and Death Valley located to the southwest of the NNSS (Winograd and Thordarson, 1975; Fenelon *et al*., 2010; Belcher and Sweetkind, 2010).

Radionuclides from underground testing in Yucca Flat, as noted previously, remain mostly in the alluvial and volcanic rocks. Where local conditions allow migration through these rocks, radionuclides are expected to move vertically downward and feed into the carbonate aquifer in the central and southern part of the basin, most likely along sets of north–south trending faults. Particle track studies for selected test locations in Yucca Flat show flow south beneath Yucca Flat, CP Basin and southwestward along the Rock Valley fault system, discharging into the Alkali Flat-Furnace Creek Ranch system (USDOE, 1997). Alternatively, flow may diverge southward across the Rock Valley fault system and terminate in the Ash Meadows discharge area (Fenelon *et al*., 2010; see Fig. 26.2). For either case, groundwater from Yucca Flat is expected to travel a minimum of 40 km from sites of underground testing before crossing the southern boundary of the NNSS.

Modeling studies of radionuclide transport in Frenchman Flat show that significant quantities of radionuclides are unlikely to reach the regional carbonate aquifer within 1,000 years. Two underground tests in the north part of the basin are located near the eastern edge of the NNSS (Fig. 26.4); radionuclide transport in the fractured volcanic aquifers from these two tests may cross the southeast boundary into Federally controlled land adjacent to the NNSS within 1,000 years (NNES, 2010a).

Preliminary estimates of the travel times through the unsaturated zone to the regional groundwater table for radionuclides from the underground

tests in the tunnel beds of Rainier Mesa and Shoshone Mountain exceed hundreds of years; radionuclide concentrations in groundwater beneath the Mesa are expected to be low. Travel time estimates to the regional groundwater table for the two tests conducted in vertical shafts in southwest Rainier Mesa are much shorter than for the tunnel bed detonations. There are multiple permissive directions of groundwater flow from Rainier Mesa: northward, southwestward beneath Pahute Mesa, or southward (Fenelon *et al.*, 2008). Southward migration of radionuclides from the Mesa areas is toward and beneath Fortymile Canyon and Jackass Flat entering into the Alkali Flat-Furnace Creek flow system (Fig. 26.2). Minimum distances of radionuclide migration from the Rainier Mesa and Shoshone Mountain underground tests to the south boundary of the NNSS are greater than 45 km for Shoshone Mountain and greater than 60 km for underground testing at Rainier Mesa.

Regional groundwater flow from testing areas of western and central Pahute Mesa is dominantly off the mesa highlands moving generally southwestward off the NNSS toward surface springs in Oasis Valley of the Oasis Valley flow system (Figs 26.2 and 26.7). Analysis of groundwater from an exploratory well located immediately outside of the NNSS boundary south of Pahute Mesa show small concentrations of tritium from underground testing, the only confirmed occurrence of local test-produced radionuclides outside of the boundaries of the NNSS.

26.4 Regulatory strategy

The regulatory strategy for the NNSS was negotiated in the 1990s as a tri-party *Federal Facility Agreement and Consent Order* (FFACO) between the DOE, the State of Nevada acting through the NDEP, and the US Department of Defense. The FFACO identifies corrective actions for historical sites of development, testing, and production of nuclear weapons and implements four stages of actions:

1. Planning of corrective action investigations.
2. Corrective action investigations consisting of site characterization of groundwater contaminated by underground testing and development of flow and transport models to forecast areas of contaminated groundwater over 1,000 years.
3. Model evaluations to test model results sufficient to develop confidence in their application to regulatory decisions required for identified corrective actions.
4. Closure in place with development of a long-term monitoring network and local implementation of institutional controls to ensure compliance.

These corrective action stages are referred to as the UGTA strategy and are based on three assumptions (USDOE, 2006). First, remedial actions to remove or stabilize subsurface radiological contaminants are neither technologically feasible nor cost effective. Second, closure in place with monitoring and institutional control of areas of groundwater contamination is the only practical corrective action. Third, the risk of contaminated groundwater is to workers, and the public. For risk to occur there must be access to contaminated groundwater. As noted in a previous section, there are no natural surface releases of contaminated groundwater on the NNSS. Exposure to contaminants requires drilling into and using groundwater from areas of present or future contaminated groundwater, actions which are restricted under current NNSS institutional control policies.

Multiple assumptions under the original FFACO agreement were used to establish the logic of the UGTA strategy. Modeling of groundwater flow and radionuclide transport is assumed to be the most effective way to identify areas of groundwater contamination over 1,000 years. An alternative approach considered during early negotiations of the FFACO agreement was simply locating monitoring wells at the perimeter of the NNSS down gradient of testing areas. The weaknesses of a monitoring-only alternative are the large number of monitoring wells required to effectively implement the strategy and uncertainty in locating monitoring wells; modeling of groundwater flow and radionuclide transport is the primary basis for developing a long-term monitoring strategy. An external peer review panel evaluated the UGTA strategy in 2001 and concluded that the corrective action steps of the UGTA strategy are logical (Institute for Regulatory Science, 2001).

The FFACO agreement assumed there would be sufficient confidence in the model results to support regulatory decisions required to complete the UGTA strategy. The regulatory metric for discriminating areas of contaminated versus non-contaminated groundwater identified in the FFACO agreement is the radiological standards of the Safe Drinking Water Act (SDWA). This is a widely applied regulatory standard used in groundwater studies by the US Environmental Protection Agency (EPA) and for many sites of environmental remediation throughout the DOE complex. For the UGTA studies, the SDWA is applied to aquifers in a remote arid desert setting, whereas the groundwater protection standards of the SDWA are normally assessed for municipal water supplies. The FFACO agreement requires an assessment of the likelihood of exceeding the radiological standards of the SDWA over 1,000 years.

26.4.1 Evolution of the regulatory strategy

Characterization and modeling studies of sites of underground testing on the NNSS were conducted after acceptance of the FFACO agreement in

the mid-1990s. However, progress was slow and multiple problems were encountered with implementing the original UGTA strategy (Marutzky *et al*., 2010). The strategy assumed sequential progress through planned characterization and modeling studies and underestimated modeling uncertainty and the importance of unanticipated scientific discoveries in characterization and modeling work. Progress, particularly progress in modeling studies, is often non-systematic with unexpected discoveries where these discoveries require rethinking of modeling approaches. Further, much of the initial modeling work for UGTA was focused on the physical and chemical processes of flow and transport rather than gaining information required to make regulatory decisions (*process*-driven modeling studies instead of *decision*-driven modeling studies). These difficulties culminated with a negative review by an external peer review panel in 1999 (IT Corporation, 1999) of the Frenchman Flat CAU data analysis and modeling studies. The panel found the studies insufficient to conclude the corrective action investigation stage of the UGTA strategy for the CAU.

The FFACO UGTA strategy was revised in 2009 (FFACO, 1996; as amended March 2010) working with NDEP to better represent the iterative nature of modeling studies, to more fully evaluate the impact of uncertainty on modeling results and to bring risk perspectives to the strategy. The original UGTA strategy was based on a standardized approach to modeling, monitoring and closure in place at all CAUs regardless of the hydrological source term and/or the proximity of testing areas to the boundaries of the NNSS. Additionally, the original strategy identified a single key regulatory decision near the end of the site characterization and model development stage (stage two of the corrective actions). If this decision was approved for an individual CAU, the studies would proceed to a 5-year proof of modeling results, followed by closure in place with implementation of a long-term monitoring network. All modeling studies would have been concluded at the end of the second corrective action stage.

Two significant changes were made in the revised strategy. First, the strategy was redesigned to be consistent with recent guidance by the National Academy of Sciences (NRC, 2007a) and the EPA (USEPA, 2009) on the use of modeling in regulatory decisions for environmental modeling. The UGTA strategy was redefined (FFACO, 1996; as amended March 2010) where the emphasis and culmination of the second stage was based on adequacy of model development. The third stage was redefined as a model evaluation stage, where model results are tested to build confidence that the model results can be used for the intended regulatory decision. The fourth stage of the strategy was largely unchanged, an emphasis on CAU closure in place and implementation of a long-term monitoring network.

26.4.2 Iterative modeling strategy and uncertainty

The second change in the strategy was designed to better represent the pragmatism of an iterative modeling approach focused on quantifying and attempting to reduce uncertainty sufficient to support regulatory decisions. Multiple decision points were added between NSO and NDEP at critical steps in the overall progression of UGTA studies. Each decision represents a juncture between continuing forward in the strategy progression or looping back (iterating) through studies. For example, a decision point was added at the end of development of a flow and transport model in the second stage to assess whether the data and model results are adequate. If both are judged adequate, the studies proceed to an external peer review. If either the data or model results are judged inadequate, the studies return to additional site characterization, refined modeling studies, and sensitivity and uncertainty analysis, all essential parts of an iterative modeling cycle.

Uncertainty in UGTA studies, particularly modeling uncertainty, was also reassessed in the strategy revisions. The multiple components of uncertainty in modeling studies are divided into statistical and structural uncertainty following guidelines established in the uncertainty literature (Morgan and Henrion, 1990; Krupnick *et al.*, 2006). Statistical uncertainty includes knowledge uncertainty and variability as a subset of knowledge uncertainty; structural uncertainty refers to model, conceptual model, and decision and regulatory uncertainty.

Reassessment of uncertainty in UGTA studies led to changes in both the approach and output of modeling studies. Modeling under the original strategy emphasized development of a preferred model of groundwater flow and radionuclide transport. However, the external peer review of the modeling studies for Frenchman Flat (IT Corporation, 1999) concluded that a single model result did not adequately represent the full range of potential model responses. The revisions in the UGTA strategy are designed to emphasize development of multiple alternative model responses that represent a spectrum of permissive combinations of model output using multiple alternative models of the hydrologic conditions and geologic setting of flow and transport in the NNSS.

26.4.3 Risk-informed perspectives

A risk-informed perspective was added to the revised UGTA strategy, recognizing the twofold nature of the project goals. The first essential goal is to complete a sufficient level of characterization and modeling studies to establish a fundamental understanding of the processes of release and transport of test-produced radionuclides in groundwater. Second, this

knowledge is applied to each CAU to identify the risk of radionuclide contamination to the public. Risk in this context is the likelihood and consequences of public exposure to contaminated groundwater and is mitigated by two factors. The first is natural attenuation or intrinsic remediation, the operation of natural processes that can reduce the concentration of a contaminant in groundwater (National Research Council, 2007a). For groundwater flow at the NNSS, natural attenuation relies on the processes of dispersion, dilution, radionuclide retardation and radioactive decay to reduce the concentration of radionuclides in groundwater from their concentrations near test cavities. Second, access to contaminated groundwater is required to complete the pathway to public exposure scenarios, the consequences portion of the risk definition. Public access to groundwater on the NNSS is restricted by the current institutional control policies. Assuming continuity of these policies, the likelihood of public access to contaminated groundwater is greatest where there is the potential for migration of radionuclides beyond the NNSS boundaries.

As noted previously, the approach to assessing the likelihood of the hazard part of the risk definition for radionuclide contamination of groundwater is developing probabilistic maps of exceeding the SDWA as specified in the FFACO agreement. The consequences of groundwater contamination from underground testing are currently controlled through implementation of worker safety protocols with respect to accessing contaminated groundwater and maintaining restrictions on public access to the NNSS. The uncertainties in these controls are the effectiveness and duration of active institutional control of the NNSS and the ability of NNSA/NSO to establish and maintain institutional controls for areas of groundwater contamination that extend off the boundaries of the NNSS.

Fig. 26.3 shows the location of sites of past underground testing on the NNSS for the major CAUs, and the expected directions of groundwater flow and radionuclide transport are shown in Fig. 26.2. The highest source term by activity in Curies is the Pahute Mesa CAU which contains less than 10% of the underground tests but more than 60% of the radiological source term (8.0×10^7 curies). The Yucca Flat CAU includes about 82% of the underground tests and 37% of the radiological inventory (5.1×10^7 curies). Slightly over 7% of the underground tests on the NNSS were conducted at the Rainier Mesa/Shoshone Mountain CAU which includes 0.7% of the radiological source term (8.9×10^5 curies). Finally, 10 underground tests were detonated at the Frenchman Flat CAU and these tests equal about 0.14% of the radiological source term (1.9×10^5 curies).

Comparison of Fig. 26.3 and the above cited distributions of the radiological source term by CAU provide important risk perspectives. The Pahute Mesa CAU contains the highest underground inventory and the greatest potential for contaminant migration off of the NNSS. As noted previously,

tritium contamination has already been detected in groundwater just south of the NNSS boundaries in western Pahute Mesa. By virtue of the high inventory and high likelihood of migration off of the NNSS, the Pahute Mesa CAU provides the greatest risk to the public. The Frenchman Flat CAU has the lowest inventory of the UGTA CAUs but the results of the transport modeling indicate a fair potential for offsite migration of radionuclides at the southeast boundary of the NNSS (Fig. 26.4). The Yucca Flat CAU includes the highest number of underground tests and a relatively high inventory, but sites of underground testing are more than 40 km away from the southern boundaries of the NNSS. Finally, the Rainier Mesa/Shoshone Mountain CAU has both a relatively small inventory and a very long expected distance of transport of radionuclides to the southern boundary of the NNSS. From a risk perspective, it is the least hazardous of the testing areas on the NNSS.

26.5 Future trends

The challenge for UGTA is to develop acceptable numerical models of long-term migration of radionuclides produced during underground testing of nuclear weapons. Contrary to many environmental contamination problems, the time and location of individual tests that produced the contamination are known and the inventory of residual radionuclides is established within the constraints of unclassified testing information (Bowen *et al.*, 2001). What is uncertain is the portion of the radionuclides that are released to groundwater (hydrological source term), and the details of radionuclide transport with groundwater flow over time. Providing this information requires development of predictive models of the long-term behavior of hydrogeological systems, a pressing societal need for a range of environmental problems (Tsang, 2005). These types of models are plagued by traditional problems in the hydrological sciences: the heterogeneity of the spatial setting of transport, the long timescales for predicting processes of flow and transport, and the problems and limitations in obtaining adequate subsurface data for developing acceptable numerical models. The traditional approach of developing a 'best predictor' model or even a 'conservative' model has been shown to be flawed (Winograd, 1990; Beven, 1993, 2000; Beven and Freer, 2001; Bredehoft, 2003; Macfarlane, 2007). The model structures of 3-D contaminant transport problems are complex, and the parameter sets required to populate the spatial domain of models are nearly always inadequate. Modeling problems of radionuclide transport in complex geological settings are always data limited, uncertainty dominated, and there are multiple sets of model and data structures that can provide acceptable simulations which honor data constraints. Independent data to test model results are rarely available and it is very difficult to decide whether

model results are reasonable representations of complex natural systems (Oreskes and Belitz, 2001).

Clearly, alternative approaches to developing and applying modeling information are required for supporting effective regulatory decision making for assessments of radionuclide migration in complex hydrogeologic settings. These approaches require a shift from over reliance on models as predictive tools to the recognition that models are information tools which aid decision making, where the decisions recognize the uncertainty in model results (NRC, 2007b). This approach replaces the outdated concept of model validation (Oreskes *et al.*, 1994) with a more relevant process of model evaluation that attempts to determine whether a model and model results are sufficiently useful to support the required regulatory decisions.

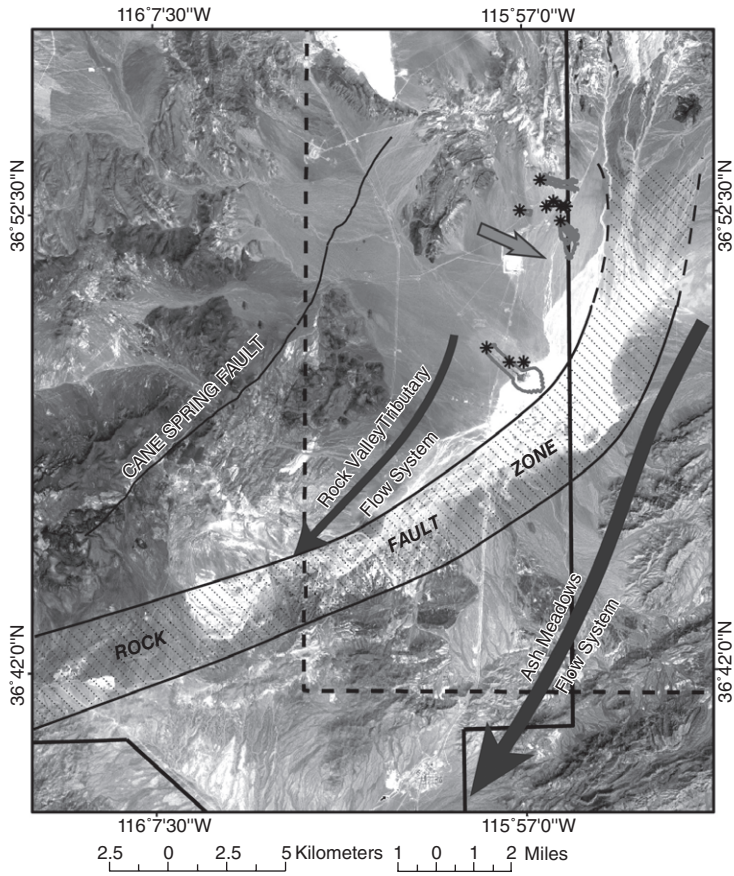
26.5.1 Frenchman Flat modeling studies

These final sections overview the results of flow and transport modeling studies and assessments of uncertainty for the Frenchman Flat CAU, the most developed of the CAU studies on the NNSS. There are three dominant features of all conceptual models of the Frenchman Flat basin (Fig. 26.4):

1. the high hydraulic heads in the CP basin northwest of Frenchman Flat (over 100 m higher heads than the Frenchman Flat basin; see Fig. 26.4),
2. the semi-perched condition of groundwater in the alluvial and volcanic aquifers with higher heads in these aquifers than the regional LCA,
3. the southeastward thinning of the volcanic section beneath the basin across Frenchman Flat.

These combined features support two inferential observations for the basin. First, groundwater flow in the alluvial and volcanic aquifers is likely horizontal across the basin from northwest to southeast (NNES, 2010a,b). Second, there is increased leakage downward into the LCA from the alluvial and volcanic aquifers as the basal volcanic confining unit thins to the southeast and/or is offset by faults associated with the Rock Valley fault system. Particle track studies originating at locations of underground tests show southeast flow through the alluvial and volcanic aquifers changing to southwestward flow in the LCA following surface and subsurface faults associated with the basin structure (Bechtel Nevada, 2005; SNJV, 2006; NNES, 2010a,b). These observations are consistent with groundwater flow converging into and following faults of the Rock Valley fault system in southern Frenchman Flat (Fig. 26.8).

Modeling studies for the Frenchman Flat CAU combine steady state and transient source term studies, multiple alternative representations of the groundwater flow system, and probabilistic transport simulations. Source term models of radionuclide releases into groundwater were developed for



26.8 Satellite photograph of the Frenchman Flat basin on the southeast edge of the NNSS showing the major structural features of the basin and directions of groundwater flow (large black arrows: regional flow system; large gray arrow: local flow in the alluvial and volcanic aquifers). The Rock Valley fault zone is a zone of echelon faults that form the Rock Valley fault system. The asterisks mark the location of ten underground nuclear tests; three in central Frenchman Flat and seven in the north part of the basin. The solid gray lines outline the edges of contaminated groundwater defined by the 95th percentile of exceeding the radiological standards of the Safe Drinking Water Act over 1,000 years. These contaminant boundaries are small (<500 m length and for some tests in alluvium, the contaminant boundaries are smaller than the asterisk symbol marking the test locations); the contaminant boundaries are larger for two tests where the underground cavity was in or near fractured volcanic rocks (two tests in the northern area) or where a 17-year radionuclide pumping experiment discharged contaminated groundwater on the surface (one test in the central area).

two settings. First, the radiological source term for underground tests in alluvium were calibrated, for both steady-state and transient models, to observed breakthrough of radionuclides at a pumping well located 91 m from the CAMBRIC test in the water table in alluvium (Tompson *et al*., 1999; Carle *et al*., 2007). Second, two underground tests in northern Frenchman Flat were conducted above the water table in or near fractured volcanic rock, where the rock permeability and porosity is inferred to be enhanced from the effects of the test detonation (IAEA, 1998). Simplified source term models were developed for these tests that account for unsaturated and saturated flow and transport and test-induced changes in rock properties (NNES, 2010a,b).

Multiple steady state groundwater flow models were developed for the Frenchman Flat CAU (SNJV, 2006) that are calibrated to hydraulic heads and permeability data for hydrostratigraphic rock units, and attempt to account for conceptual model uncertainty. The evaluated components of conceptual (structural) model uncertainty include variability in boundary conditions and boundary fluxes, permissible alternative hydrogeological frameworks for the basin, including structure (faults and basin features), stratigraphic units within the basin, and alternative recharge models. The goal in developing flow models was not to identify a best-fit calibration or a best predictor flow model but instead to distinguish a range of alternative flow models that capture the range of variation in flow fields from parametric and structural uncertainty. This range in groundwater flow was then used in transport simulations. Statistical metrics of goodness of fit of alternative groundwater calibrations did not provide useful information for discriminating or screening groundwater flow models. Two alternative sets of data did provide useful information for categorizing results for calibrated flow models (SNJV, 2006). These include variability in particle track results, and variability in groundwater velocity and direction at test cavity locations using linear predictive uncertainty analysis from parameter estimation software (PEST; Doherty, 2007).

Monte Carlo transport simulations were conducted for underground tests at the two testing areas in Frenchman Flat (Fig. 26.4). Four flow models were combined with alternative sets of boundary conditions (boundary fluxes, hydrostratigraphic frameworks and recharge) to represent the variability in the groundwater flow field (velocity and direction of flow at the test cavity). These flow conditions were established at the underground test cavities as the initial conditions for transport simulations sampling stochastic transport parameters using a streamline-based convolution transport code (Robinson *et al*., 2011). Radionuclide concentrations for 1,000 years of transport were post-processed to develop probabilistic forecasts of exceeding the radiological requirements of the SDWA (Fig. 26.8); the boundary of this representation denotes the limits of contaminated

groundwater (contaminant boundary) defined as a 5% chance or less of exceeding the SDWA. There are two categories of contaminant boundaries: (1) small boundaries (<500 m maximum lateral distance) where the test cavity and transport are in the alluvial aquifer and (2) larger boundaries (>1600 m) where the source term and/or transport is in fractured volcanic rock. For the latter category (two underground tests), the contaminant boundaries extend slightly off the NNSS boundaries into adjacent Federal land (Fig. 26.8).

The contaminant boundaries of the central testing area of Frenchman Flat (Fig. 26.8) are complicated by two factors. First, the long-term pumping test for the CAMBRIC test discharged contaminated groundwater on the surface into a ditch that drained into the Frenchman Flat playa. Second, the discharged contaminated water in the drainage ditch infiltrated to the water table in concentrations that exceed the SDWA. This required transient models to account for the 17 years of continuous aquifer pumping and surface discharge of contaminated water and significantly extended the contaminant boundaries of the central testing area.

The contaminant boundaries depicted in Fig. 26.8 will be used for two regulatory decisions. First, the boundary geometries will be used to designate surface use restriction areas where institutional controls will be imposed to restrict all drilling to potentially contaminated groundwater. Second, the contaminant boundaries and results of subsequent monitoring studies will be used by NDEP to identify a regulatory boundary designed to protect the public and environment from exposure to contaminated groundwater. The NNSA/NSO will be required to develop a plan to mitigate potential impacts on the public, if radionuclides are detected at the regulatory boundary. The regulatory boundary has tentatively been identified as the Rock Valley fault zone at the southern end of Frenchman Flat, the expected migration pathway to public access to groundwater south of the southern boundary of the NNSS.

The transport model for the Frenchman Flat CAU was accepted by NDEP following successful external peer review of the CAU studies (Navarro-Intera, 2010). This marks the first successful completion of the model development stage under the UGTA strategy and the initiation of the model evaluation stage for the Frenchman Flat CAU (USDOE, 2011).

26.6 Acknowledgments

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27

Remote monitoring of former underground nuclear explosion sites predominantly in the former USSR

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Abstract: This chapter reviews and discusses the effects of residual features on the long-term geothermal activity in the epicentral zone of underground nuclear explosions (UGE). The thermal anomaly parameters and their connection to carrying out thermal surveys and surface thermal logging on the present day surface are determined. A remote method of measuring the thermal anomalies is proposed.

Key words: underground nuclear explosion, radioactive waste, thermal radiation, monitoring, epicentral zone.

27.1 Introduction

Worldwide, 2,054 nuclear explosions have been conducted since 1945, including 1,524 underground explosions (many explosions were carried out in groups) (Kochran *et al*., 1992; Mikhailov, 1992, 2001). The last 1,373 explosions were performed at special nuclear test sites:

- 333 explosions at Semipalatinsk and West Kazakhstan (former Soviet territory, at present the territory of the Republic of Kazakhstan),
- 39 explosions at Novaya Zemlya (Russia),
- 781 explosions in the US (Nevada),
- 3 explosions in the USA on the Island of Amchitka (Alaska) landfills,
- 13 explosions in Algeria (District Hoggar),
- 147 explosions on the islands of Mururoa and Fangataufa (France)
- 24 explosions at the Nevada test site in the US were performed by the United Kingdom,
- 24 explosions were performed at the Lop Nor test site in China,
- Miscellaneous test explosions were carried out by India (3), Pakistan (2), and North Korea (2) (Mikhailov, 2001).

Other underground nuclear explosions were carried out underground at various test sites or on the surface, but with the purpose of applying the technology of nuclear explosions for peaceful solutions of a variety of technical problems (Mikhailov, 2001; Logachev *et al*., 2001).

In the Soviet Union from 1961 to 1987, in accordance with Programme No 7 'Nuclear explosions for the national economy,' 124 industrial complexes experienced an explosion, of which a number were carried out at the Semipalatinsk test site. Outside the territory of the present-day Russia, 80 explosions were carried out in the Republic of Kazakhstan (outside the polygon), 32 in the Ukraine, two in Uzbekistan, and two in Turkmenistan. The majority of the explosions were carried out in camouflet option, i.e. without a breakthrough cavern explosion into the atmosphere, and were aimed at solving problems: seismic sensing (39), creation of industrial containers for food storage (26), working out the technology and scientific experiments (22), intensification of oil fields (21), eliminating emergency fountain (5), creating reservoirs (4), waste disposal in deep horizons (2), crushing ore (2), prevention of gas emission in coal seams (1), creating channels (1), and tailings dams (1) (Mikhailov, 2001; Israel, 1974).

Most of the explosions were carried out under difficult physical and geological conditions: in permafrost, semi-deserts, mountains, and salt formations in mining areas. Together with the explosion parameters and the monitoring information, these conditions determine the nature of residual geophysical phenomena, i.e. cleavage zones, zones of increased fracturing, changes in the permeability induced by electric and magnetic fields, thermal effects, and possible contamination with radioisotopes, which are precursors of volatile radioactive elements, increased release of radioactive radon gas, and changes in the environmental performance of the natural environment, etc.

This chapter describes the features and control areas of underground nuclear explosions and potential changes over long time periods, which allow evaluation of the state of the environment, i.e. the outward manifestation of certain physical fields on the surface.

Section 27.2 describes the basic mechanisms of the boiler cavity, pillar collapses, and the cleavage phenomena on the surface, while also summarizing the classification and spatial distribution of radioactive waste. Section 27.3 examines the long-term problematic situations that arise at the surface, in aquifers and hydrocarbon horizons in the zone of underground nuclear explosions. There are cases that require regular monitoring. Section 27.4 is devoted to describing the results of thermal imagery and ground temperature well logging in areas of underground nuclear explosions. A phenomenological model of formation and dynamics of thermal anomalies is developed. Links are made between thermal anomalies, the level of gamma background radiation, and radon releases. In Section 27.5 we propose a method using monitoring by spacecraft to measure thermal anomalies. The prospects of applying this method for global monitoring of the effects of underground nuclear explosions are determined.

27.2 Effects of the underground nuclear explosions on the environment

When a camouflet explosion occurs under high temperature (over a million degrees Kelvin) and high pressure (order of several million atmospheres), evaporation and melting of rock occurs in the region where the charge was laid, resulting in a boiler chamber having a shape similar to a three-dimensional ellipsoid. The effective radius of this cavity is 10–40 m. The cavity wall thickness is several tens of centimeters, composed of sintered layers of rock. The mass of the melt reaches 400 m at 1 kiloton of explosive power. Behind the wall cavity, as a result of the shock wave, is crushed rock. At large distances behind the wall cavity, is a region of increased fracturing. A truncated cylinder shape is formed with the upper limit in the cleavage zone impacting the surface of the Earth above the boiler cavity zone where increased fracturing occurs (Israel, 1974).

Over time, gravity causes the melt to flow down from the top and side walls of the cavity to its lower part, forming a lens of melt. After a further decrease in temperature, the melt passes into a solid phase and is partially or completely embedded with fragments of rock up to a height of a few meters from the bottom of the cavity. In this case, the bottom layer of the fractured rock pile covers the lens of melt. The array of the rock above the boiler cavity has been destroyed and eventually starts to sink down to form a pillar collapse. This process partially reverses the expansion of soil and rock mechanical faults caused by the shock wave, but also lowers the gas pressure in the cavity that was formed. Since the diameter of the column collapses the diameter of the boiler cavity, the cave only partially fills the cavity, forming one or more hollow zones located closer to the surface. The pillar collapse has very high moisture and gas permeability. The associated, filtration coefficient is hundreds of meters per day, and the coefficient of loosening of pillar collapses, defined by the ratio of porosity before and after the explosion, reaches 0.73–0.85. At the same time, the lateral border pillar collapses and is clearly separated from the solid undisturbed rock. At the point of contact, the lateral border pillar collapses along with the adjacent undisturbed rock to form a peeled zone with permeability greater than the permeability of the collapsed column. At the ground surface above the explosion zone epicenter cleavage phenomena were observed. These took the form of swelling or rock subsidence depending on the exact nature of the explosion. Often crushed rocks are observed on the rock – similar arrays in the cleavage zone.

Because of the complexity of nuclear processes, a range of radionuclides are released in the explosion, which are deposited mainly in the cavity of the explosion. High-melting products are concentrated mainly in the lens

of the melt and are mostly fissile nuclides of uranium and plutonium, fission fragments and neutron-activated elements of the charger and breeder. In the column collapse and fracture zone, volatile compounds such as plutonium and polonium are concentrated, as well as the radionuclides strontium, cesium, lanthanum, etc. (Israel, 1974).

27.3 Problems as a consequence of underground nuclear tests

During surveys of the territories of Semipalatinsk nuclear test site by American satellites NOAA-14 and NOAA-15, experts at the National Nuclear Centre of Kazakhstan detected large-scale surface temperature changes (Zakarin *et al*., 1997; Sultangazov *et al*., 1997). Their findings indicated the presence of a regional thermal anomaly with a surplus temperature of about 10°C in an area which was over 20,000 km^2, i.e., the entire area of the landfill including the sites of Degelen and Balapan. The presence of such a thermal anomaly was assured to be associated with increased activity of the earth surface and the active mechanism of 'smoldering' reactions of nuclear fission. It is hypothesized that, under the influence of gamma radiation in the atmospheric boundary layer, reactions occur that result in a certain part of the oxygen being converted into ozone (Melent'ev and Velikhanov, 2003). Since ozone is heavier than air, it is concentrated at the surface of the Earth and, having been an active oxygenator, produces detrimental effects on biological systems. This effect is confirmed by the images obtained from satellites: there is practically no vegetation in the places that experienced these higher temperatures. Publications on this issue are the subject of much scientific debate. It is clear that the parts of the Earth's surface exposed to nuclear explosions should be looked at in more detail to examine the structure of the thermal field at the landfill, in order to draw attention to the complex combination of natural conditions and radiation effects, taking into account the low spatial resolution of the apparatus of NOAA satellites.

In addition, these influences are manifested at the ground surface (under certain conditions they can be observed visually, such as when snow melts in the warmer parts of the area). However, all processes associated primarily with the underground migration of radioactive products in the aqueous and hydrocarbon layers (including the partitioning of radioactive products in the area of the boiler cavity from a melt solution, and their contamination of surface and groundwater) and changes in the hydrological regime of aquifers are hidden from the naked eye.

The articles by Kiryukhina and Shahidzhanov (2003) and Bakharev *et al*. (2002) specifically note the possible effects of long-term exposure of elements of the cavity to radionuclides and the post-explosion collapse of

aquifers after different times. In this case, additional man-made caverns and aquifers contaminated with radionuclides may produce an ever-expanding contaminated area in concert with the natural aquifer system. It is noted that the radiation risk can increase substantially if the boiler starts to accumulate karst cavities or other water, that interacts with calcium oxide which can serve as a basis for the formation of liquid radioactive brine (calcium hydroxide), which is able to penetrate sufficiently large distances, up to the upper layers of aquifers. With technological processes occurring near such cavities, the removal of radioactive material to the surface should not be excluded. In limestone-containing rocks, these processes can be exacerbated by the fact that it is likely that the crushed pile containing calcium oxide and carbon dioxide will expand and will be distributed through permeable systems and brought to the surface through increased fracturing.

Observations on the migration of radioactive products from underground nuclear explosions carried out in permafrost conditions have been described by Golubov *et al*. (2003) and Kozhukhov and Kukushkin (2003). The distribution of radon, tritium, strontium and other radionuclide contents in the water, and gamma radiation in the vicinity of the explosion 'Crystal', carried out in 1974 in Yakutia near the diamond-mining quarry known as 'Udachnyi', were studied. Measurements were carried out from the epicenter to the quarry (about 5 km) and showed the following:

1. The level of gamma radiation ranged from 9 to 14 micro-R/h, i.e. it did not exceed natural background levels when the whole area was surveyed.
2. The volume of the radon activity in the epicenter, at a distance of 2.5 km, ranged from 400–500 to 1,300–1,400 Bq/m^3.
3. In the area of the quarry, the radon content was 200–700 Bq/m^3, suggesting that the rate of migration of radon in the local soil is low.
4. There is increased concentration of tritium to 220 Bq/l in the epicenter of the explosion.
5. Concentrations of radioactive carbon and strontium in the drained brines on the side quarry of 'Udachnyi' are on average 2–3 times higher than the corresponding concentrations in groundwater from technological wells close to the background level.
6. It cannot be excluded that the permeability of permafrost rocks in this area caused the working quarry horizons to drop to a much greater depth than that of the cavity created by the nuclear explosion, thereby promoting the drainage of underground brines in the vicinity of the cavity wall of a quarry with the formation of the network of flooded cracks with dissolved radioactive products.

Thus, according to Bakharev *et al*. (2002), each underground nuclear explosion site creates a self-generating uncontrolled dumping of radioactive

products into the environment that can have a permanent impact on nature and mankind and, therefore, should be regarded as a functioning 'radiation-dangerous' object. Evaluation of radiation and ecological safety in this case is connected with the prediction of the secondary impacts of the residual effects of an explosion on the environment and should be based analysis of situations that could lead to further dissemination and redistribution of the radioactive products.

27.4 Thermal anomalies as informative signs of underground nuclear explosions

Investigation of residual effects from peaceful explosions is a laborious and expensive task, requiring the creation of special missions with the appropriate hardware and monitoring equipment including vehicles, staffed by highly qualified scientific and technical personnel. For example, to study the thermal fields, among other things, requires manned aircraft. It is considerably more convenient to study the geophysical implications and methods of their control at test ranges where a developed technological infrastructure and trained personnel with the necessary qualifications exist to ensure that the results of these studies for relevant peaceful uses of nuclear explosions are adequate. Therefore, a significant part of the material in this section is based on the results of experiments conducted at the Semipalatinsk nuclear proving ground.

The majority of the surveyed explosions took place at the Degelen mountain range, located near the Kalba-Chingiz deep fault. This complex, mostly granite, volcanic and volcanic-sedimentary rocks, forms a large structure with a diameter of about 30 km. Intrusive rocks are interspersed in the form of individual granite-like bodies of relatively small size. A smaller part of the surveyed explosions were in the area of the test site Balapan located close to the eastern border of the landfill. Geologically, much of it is placed in the Zaisan folded region. A latitudinal piece of the Kalba-Chingiz deep fault, which separates this area from Chingiz-Tarbagatai, runs almost along the southern border of the latter. The depth of the water table is 200–400 m. The entire area is characterized by a homogeneous filler surface, folded eluvial sands of 4–6 m, or dense clays (Busygin and Andreev 2004).

Climatic conditions at Semipalatinsk are sharply continental with an average temperature of about +1°C. Summer is hot and dry with temperatures up to +40°C. Autumn and spring are cloudy and cold with average temperatures not higher than +7°C. The exception is May, when it is warm and clear. Winter is cold with little snow and with temperatures as low as –40°C. These geological and climatic characteristics of the area determine the conditions of conservation of thermal lesions in the rocks, the formation

of thermal anomalies on the ground surface, and the possibility of their detection.

The first results of the thermal regime created by the underground explosions (UGE) on the ground surface were obtained in the late 1980s and were published in a series of papers by Busygin *et al*. (1999) and Busygin and Andreev (2004). First ring-shaped forms were discovered covering the cleavage zone of the UGE as they were luminous in the infrared spectrum. The physics of these phenomena remains unclear. The formulation and solution of rigorous mathematical tasks was required to describe the processes of heat transfer and gas flow. However, a comprehensive package of initial data and a set of direct measurements of temperature and air flow in the cavity and the Earth's surface, made in a wide range of temporary, geometric, and meteorological conditions, was also required.

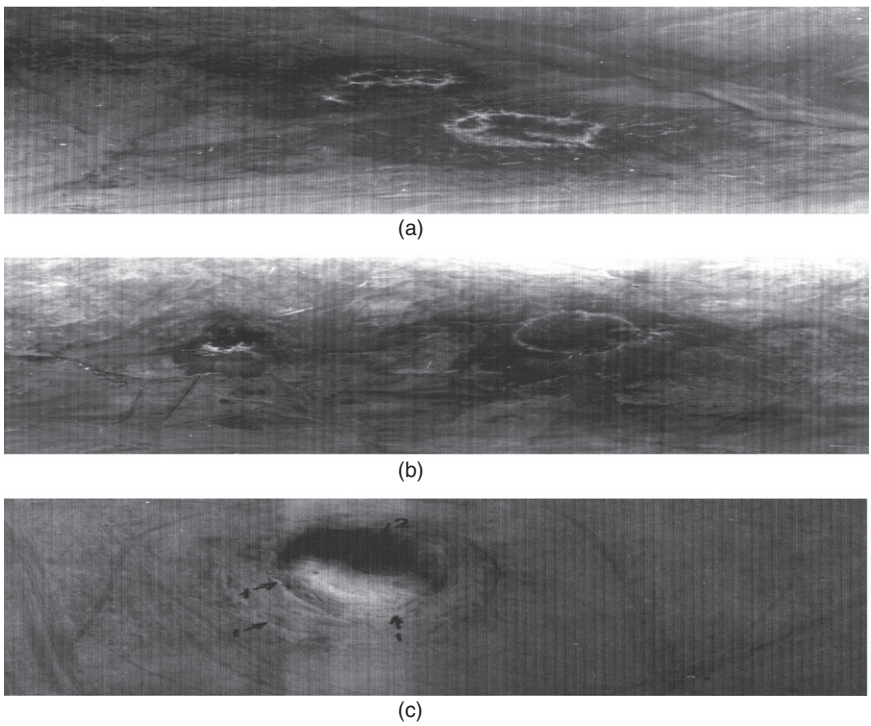
Review of materials on the sprung hole of a UGE shows that for many years they have a high internal temperature, slowly decreasing over time (Israel, 1974; Taylar, 1973). Results for the domestic UGE show that the average air temperature in the boiler cavities of the explosion conducted more than 10 years before, is 30–50°C, i.e., the boiler cavities of UGEs are long-term sources of heat.

It follows from Section 27.2 that the boiler cavity after the UGE is not absolutely airtight. The presence of anthropogenic influences, fracture zones, column collapses and other tectonic features makes the contents of the boiler cavity available for air transport and, consequently, for the removal of heat and gases present in the cavity to come to the surface. To control the intensity and configuration of thermal anomalies on the ground surface, the method of heat shot is employed from onboard aircraft, using the 'Volcano' thermal imaging equipment which is modified with a unit controlling the film transport rate, which requires a flight height range of 200–3,500 m above the surface. The method of optical-and-mechanical scanning was used in the direction perpendicular to the direction of travel of the thermal imager in the aircraft. The flights carried out tasks over the examined area, and the height of the flight was supposed to provide the required coverage.

The optical part of the recording apparatus was a cooled infrared radiometer with a sensitivity of 8–14 microns. The sensitive nature of the equipment required that it be placed in a hanging gondola on the outer side of the fuselage of the carrier, which eliminated the effects of the aircraft glass windows. Along with the heat-sensing aerial photography conducted in the visible spectrum which allowed detailed information about the surrounding landscape to be obtained, there was a need to decrypt the thermal images and a need to accurately reference the area of the thermal objects. In this way (Busygin and Andreev, 2004), more than 50 UGE were examined during the period from 1 to 26 after the date of the initial measurement.

Almost all of the surveys performed on the ground surface in the epicentral area were observed to be ring-shaped or curved thermal structures, covering the cleavage zone of the explosion. The typical form of these structures is shown in Fig. 27.1.

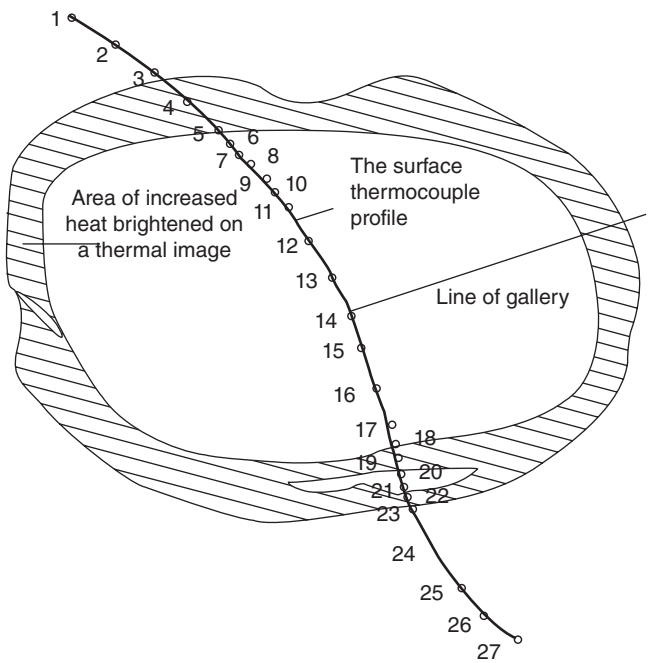
To validate the existence of thermal anomalies, as long-term residual processes occurring in boiler UGE cavities, investigations were carried out in two directions. The first set of investigations was connected with the hypothesis of uneven solar heating of the soil due to the different solar exposure of mountain slopes and micro-relief. To this end, a loop of night and pre-dawn measurements in autumn and winter under cloudy conditions with zero duration of sunshine and little difference in day and night values of air temperature were performed. The results confirmed the presence of ring-shaped thermal anomalies. Indirectly, the role of solar warming from the thermal anomalies is refuted, as solar radiation during the cold season could 'warm up' only one side of the failure cone and warming was found in these ring-shaped patterns.



27.1 Typical view of a thermal anomaly caused by an underground nuclear explosion on the surface during daytime (Busygin and Andreev, 2004): (a) and (b) explosion in gallery; (c) explosion in shaft.

The second set of investigations was conducted to test the binding of thermal anomalies on the ground surface to a picture of the local actions of UGE. The problem was solved using ground-temperature well-logging methods in the area of the thermal anomaly tied to the locality on the thermal image. Measurements of ground surface temperature were made with copper wire resistance thermocouples (temperature sensors); the standard error did not exceed 0.2–0.4°C. For the measurements of each thermal anomaly, one or two measurement lines were created. Not less than 20 sensors were placed along a cable line at a distance of about 5 m from each other (Fig. 27.2). Measurement lines were located on the ground around the diameters of circles covering a cleavage zone. The sensors are protected from direct solar radiation by special shields. The true value of the measured temperature T was calculated for each sensor separately after adjusting for the actual impedance of the line. Each cycle of measurements was carried out for three days with interval readings after 2 hours. The duration of one data point on one line does not exceed 10 minutes.

Figure 27.3 shows the typical spatial distribution of temperature for the autumn–winter period for the profile of the location of temperature sensors

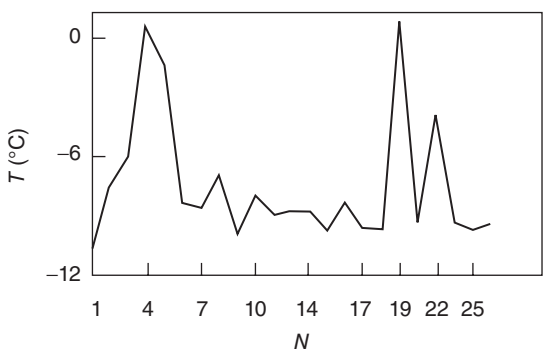


27.2 Scheme of the thermocouple placement on a thermal anomaly (Busygin *et al.,* 1999): solid curve is the surface measurement line; the circles with numbers are the numbered thermocouples.

shown in Fig. 27.2. Distances between sensors are marked as the abscissa on a proportional scale. It is evident that sensors located in a highlighted strip correspond to higher values of ground temperature compared with background values of temperature (about –9°C). The excess temperature reaches 8–10°C.

Figure 27.3 also shows that the gases exiting to the Earth's surface have a temperature lower than the rock at the charge depth (6–8°C throughout the year). This has two causes. First, the cold-season air passing through an explosion cavity that is 20–40°C did not have sufficient time to warm up due to the high velocities of the air masses. Second, due to a lack of integrity arising from formation of a large number of deep cracks, there is deeper cooling of the rocks in the array, which significantly increases the contact area of the exhaust air from the cooled rock. To confirm the fact that the removal of heated air instead of air at the natural temperature of the boiler at the depth of the cavity was examined, a peaceful UGE was conducted in Kalmykia (Russia) in the warm season, i.e. at a background temperature of 21–23°C (Granberg *et al*., 1997). Temperature thermal anomalies for it reached 28–34°C, which certainly indicates the presence of an artificial heat source from the UGE.

In parallel with the temperature well logging, estimates of the geometric dimensions of thermal anomalies were made. It was shown that a sufficiently broad energy spectrum at the depths of the UGE gives the maximum radius of the thermal anomalies which varies from 80 to 250 m, while the width of the thermal ring varies from 20 to 60 m. It was not possible to establish the full duration of thermal anomalies, as over a nearly ten-year period, their thermal anomalies remained virtually unchanged. For the UGE held in galleries, the largest fixed term for thermal anomalies at the time they could be observed was 25–26 years and for UGEs conducted in wells it was 16–18 years.

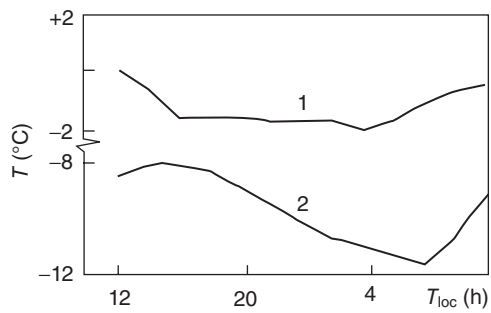


27.3 Temperature distribution on the surface measurement profile (Busygin *et al.,* 1999): the numbers *N* indicate the thermocouple numbers of the profile shown in Fig. 27.2.

It is certainly interesting to study daily and seasonal measurements of the thermal effects of UGEs at individual sites. Diurnal temperature variation, obtained by simultaneous measurements on a strip heater removed from the UGE and from the undamaged section of the Earth's surface, averaged over 48 experiments (October–November), is shown in Fig. 27.4 (here t_{loc} = local time). It can be seen that the thermal effect at the UGE site was observed continuously for days in the field, according to the thermal image, due to removal of heat from the air cavity (line 1). Characteristically, the temperature fluctuations during a day in the field of thermal anomalies are about 1°C, while for the damaged portion of the UGE, site surface peak-temperature reaches 4°C.

Significant differences are observed in the form of plots of temperature versus time for undisturbed and disturbed UGE sites. For undisturbed sites, the temperature dependence is very ordinary, without thermal anomalies in the afternoon heating and only minimum temperature anomalies at 7–8 a.m. All this also suggests that the observed thermal anomalies are not the result of solar heating of the Earth's surface and that the surface albedo changes under the influence of the UGE.

Seasonal temperature variation, in contrast to the daily temperature variation, was studied the least. In particular, during the warmer months there have been instances when the UGEs conducted in groups decreased by 2–3°C in the cleavage zone compared with the background temperature. To explain such phenomena, a phenomenological model for the formation and dynamics of thermal anomalies based on the principles of 'heating effect' was proposed. Its essence lies in the fact that the movement of air through the heated boiler cavity occurs by gas convection, and the direction of motion can be either from the portal tunnel up through tectonic faults in the epicentral area, or vice versa. From the equation for the depression



27.4 Diurnal surface temperature variation in area of thermal anomaly (Busygin and Andreev, 2004): 1, undisturbed area; 2, heat efflux area determined on photograph.

thrust air $h_e = A(t_B - t_H)$, where A is a coefficient for atmospheric parameters and channel exhalation of air; t_H is outside air temperature; and t_B is averaged over the profile of raising the air temperature inside the rock, it is evident that the magnitude of depression is proportional to the temperature difference outside and passing along the tectonic disturbance of air, and the direction of motion is determined by the sign of this difference. If the temperature t_B is calculated by using the empirical formula $t_B = 1.1(t_p - 6)/H + 6$ (Busygin *et al.*, 1999), where t_p is air temperature in the boiler cavity, and H is the reduced depth of the UGE, we can obtain approximate values of the external temperature of a UGE site, for which one should observe a positive depression ($h_e > 0$). For example, for an explosion with the yield 1 kt, warhead detonation depth $H = 100$ m, a positive depression is observed when the outside temperature does not exceed 16°C if the air temperature in the cavity is 100°C. If the temperature in the cavity decreases to 20°C, the boundary outside temperature decreases to 7–7.5°C.

The estimates given are quite approximate until a full-scale experiment can be carried out with monitored directions of transport and air flow to the outside air temperature. It should be noted that the direct measurement of air movements is possible only in the portal tunnel. In the area of the cleavage phenomena, as mentioned above, anemometric measurements are difficult due to the complexity of micro-relief areas and the inability to visually determine the position of the majority of cracks, which serve as conduits to move the air.

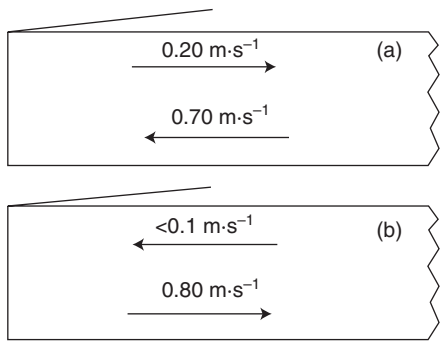
Air mass velocity was measured using an anemometer at a distance of 40–50 m from the tunnel portal. The direction of air mass movement is determined by the deviation of the flame or the direction of motion of smoke from burning smoke grenades (at speeds below 0.2 m/s). The measurements were performed at two points located at the 'top' and 'bottom' gallery. In each session, measurements of velocity were carried out at least three times for a duration of 10 s. By measuring the mean values taken for air velocity at the point of measurement, the air flow can be calculated. Results are summarized in Table 27.1 which indicate the following:

1. In the warm season, as shown in Fig. 27.5, the air velocity at the 'bottom' gallery is directed outward and is 0.25–0.70 m/s and the speed at the 'top' is directed inward and has much lower values: <0.1–0.2 m/s, i.e. the bulk of the air moves toward the portal.
2. The measured air flow, at the same time, is in the range 450–700 m^3/hr.
3. In the cold period at temperatures below ambient, the movement of air masses in the tunnel goes toward the explosion cavity. In this case, with decreasing temperature, the velocity of the air inside the tunnel increases. Air consumption compared to a warm season is reduced and varies between 180 and 350 m^3/h.

Table 27.1 Direction velocity and flow rate in gallery

Temperature of external air (°C)	Direction of air flow	Flow velocity at 'bottom' and 'top' (m/s)	Flow rate (m^3/h)
22.4	To gantry	0.70–0.22	707
20.4	To gantry	0.75–0.20	495
17.0	To gantry	0.60–0.20	475
17.3	To gantry	0.65–<0.10	475
16.2	To gantry	0.73–0.10	466
16.2	To gantry	0.22–0.20	454
19.8	To gantry	0.70–0.10	466
21.0	To gantry	0.70–0.10	466
21.8	To gantry	0.72–0.20	705
0.5	To cavity	<0.10–0.30	314
–5.4	To cavity	<0.10–0.30	348
–6.0	To cavity	<0.10–0.20	180
–9.0	To cavity	0.10–0.30	296
–7.0	To cavity	<0.10–0.20	226
–7.4	To cavity	<0.10–0.40	226
–12.2	To cavity	<0.20–0.80	–
–14.0	To cavity	<0.20–0.90	–

Source: Busygin et al. (1999).



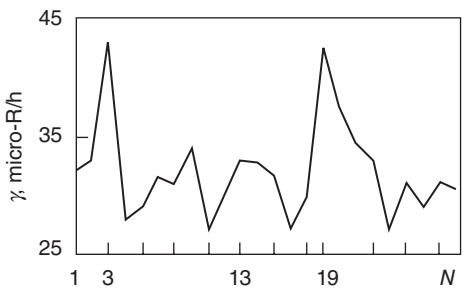
27.5 Typical variations in the velocity and direction of the air flow in galleries (Busygin *et al.*, 1999): (a) warm period of year; (b) cold period.

The experimental results qualitatively confirm the adequacy of the proposed model to real processes. It should be noted that in wells, a high temperature persists for much longer than in galleries because the heat loss occurs only due to natural convection (i.e., there is no is 'stove' effect). According to field measurements at Semipalatinsk site, the temperatures in the wells have decreased to 42–45°C some 6 years after the explosion, while in the galleries the temperature has been observed for 1.5–2 years.

Along with air, radionuclide products are transported to the Earth's surface. Direct measurements of the exposure dose on the profile of thermal anomalies have shown that in this case the radiation levels are 3–5 times higher than natural background levels (Fig. 27.6). Comparison of temperature and gamma-radiation curves indicates a high degree of correlation of these two processes. The distribution of activity and concentration of radon behaves similarly. In the location of the thermal anomaly, the volume of radon activity is 80–100 Bq/m^3. At the same time over the epicenter of the explosion, the natural background of ionizing radiation remains: 5–10 micro-R/h for gamma-rays and 30–40 Bq/m^3 for radon.

27.5 Space monitoring of thermal anomalies and prospects for its application

The method of thermal imagery is considered to be one of the most modern and effective methods of scanning terrestrial objects. For successful detection and identification of the UGE-controlled objects, such imagery requires knowledge of the spectral characteristics of radiation, weakening of the pathway of the working range of wavelengths, as well as the characteristics and capabilities of equipment in the temperature and spatial resolution of a UGE. Efficiency of detection of thermal anomalies from space can be increased by multispectral imaging including the use of the visible spectrum that provides a higher quality of decoding images and binds heat-radiating objects to the terrain. Low-orbiting satellites or space stations may be used as carriers of the recording apparatus. Although satellites and space stations both have long orbital paths of observation, the long-term survival of thermal anomalies allows them to receive and store information on the same site area due to the lack of restrictions in the number of times they can review the UGE sites.



27.6 The radiation background on the surface measurement profile (Busygin and Andreev, 2004): the numbers *N* indicate the numbers of the thermocouples in Fig. 27.2.

Transfer of infrared radiation on the 'Earth-Space' tracks took place in the spectral range from 8 to 14 microns (comparative assessments in some cases took into account the adjacent region of the spectrum). The radiation detector was focused on the thermal anomaly, with an ideal spectral characteristic in the range of wavelengths, located at the altitude of the spacecraft orbit equal to 300 km. The zenith angle of sight ranged from 0° to 80°. The distributions of basic meteorological parameters are used to characterize the atmospheric conditions in cloud-free atmosphere within their natural variability in the warm and cold periods of the year (McClatchey *et al.*, 1972). Gas models include vertical profiles of pressure, temperature, density, and the amount of water vapor, carbon dioxide and ozone as meteorological parameters, to a greater degree of influence on the transfer of radiant energy in this spectral range. Aerosol atmospheric models include a set of basic types of aerosol particles (dust, water soluble, water-dust, soot particles, acid aerosols, volcanic dust), the vertical distribution of their concentrations, the spectral values of volume extinction σ, the scattering β, and absorption δ coefficients for local and continental aerosol types.

A quantity that must be determined is the extinction coefficient E (flux density of radiation from a source of unit power) as a function of orbital altitude H, the zenith angle of sight v, complex of meteorological parameters M, the spectral range $\Delta\lambda$ and calculated as a linear functional:

$$E(v, \Delta\lambda) = T_{sc}^{M}(v, \Delta\lambda) \cdot T_{ab}^{M}(v, \Delta\lambda) \cdot T_{ex}^{a}(v, M, \Delta\lambda) \cdot T_{g}(H, v), \qquad [27.1]$$

where T_{sc}^{M} is the attenuation due to the weakening of the molecular scattering of radiation, T_{ab}^{M} is the weakening due to molecular (gas) absorption of radiation, T_{ex}^{a} is the radiation attenuation due to scattering and absorption by aerosol and T_g is the radiation attenuation due to geometrical factors.

The function in Eq. [27.1] is calculated from the following relations:

$$T_{sc}^{M} = \frac{1}{\Delta\lambda} \int_{\Delta\lambda} \exp\left[-\frac{0.0119}{\cos v} \left(\frac{\lambda_0}{\lambda} \right)^4 \right] d\lambda; \qquad [27.2]$$

$$T_{ab}^{M} = \prod_{j=1}^{m} P_{\Delta\lambda}^{j}(v, \Delta\lambda); \qquad [27.3]$$

$$T_{ex}^{a} = \frac{1}{\Delta\lambda} \sum_{i=1}^{n} \Delta\lambda_i \exp\left\{ -\frac{1}{\cos v} \int_{0}^{H} [\beta_{ex,\Delta\lambda_i}(z) + \delta_{ex,\Delta\lambda_i}(z)] dz \right\}; \qquad [27.4]$$

$$T_g = \frac{1}{4\pi R^2(H, v)}. \qquad [27.5]$$

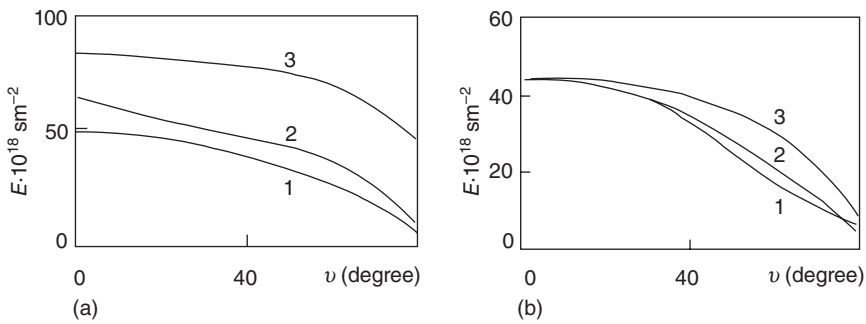
Here z is the current height above the Earth, $P_{\Delta\lambda}$ is the transmission function of the atmospheric gases, $\lambda_0 = 0.55$ micrometers, H is the ceiling of the

atmosphere equal to 80 km, and R is a distance from the source to the receiver.

The greatest difficulty in calculating the factors given in Eqs [27.2–27.5] is the calculation of the transmission functions in Eq. [27.3]. The methodology for calculating the transmission functions is chosen in accordance with the work of McClatchey *et al.* (1972) and allows determination of the attenuation due to a selective absorption of atmospheric gases and water vapor continuum absorption. Calculations of extinction coefficient are presented in Fig. 27.7 as the E function of the zenith angle of sight v. The figure shows that the influence of aerosol extinction and molecular scattering is much weaker than the gas absorption. This explains the higher values of spectral transmittance in the cold season compared to the warm. From the graphs it follows also that a change in viewing angle from 0° to 80° for all weather conditions and satellite altitudes can be incorporated in a single change to the extinction coefficient E.

The energy flux density value of the extinction coefficient E should be multiplied by the flux of the intrinsic radiation source in the corresponding intervals. Self-radiation of the UGE thermal anomaly can be approximately estimated as gray-body radiation with a surface area equal to the square of light, which manifests itself in the thermal image. Assuming that the emissive capacity $F_{\Delta\lambda}(T)$ of the thermal anomaly is constant throughout the area, for typical sizes and temperatures, radiation flux density on orbit with a height of 300 km is in the spectral range 8–14 micrometers which is quite high for the infrared radiation quantity of about 10^{-9} W cm^{-2}.

It should be borne in mind that detection of thermal anomalies against the background of the outgoing radiation from the Earth and the atmosphere depends on the response of the radiometer receiving element to temperature change, i.e. on the temperature contrast $\Delta T = T - T_{bg}$ or to change in the radiation flux density, i.e. on the energy contrast



27.7 Extinction coefficient depending on sight angle for high (a) and low (b) transparency of atmosphere: 1, $\Delta\lambda$ = 4–5 micrometers; 2, 8–10 micrometers; 3, 10–12 micrometers.

$K = (\Phi - \Phi_{bg})/(\Phi + \Phi_{bg})$. Here T and T_{bg} are the temperature of the thermal anomaly and background, and Φ and Φ_{bg} are the relevant flux density in the orbit of the satellite/space station, provided that the spatial resolution of the detectors is close enough to the size of the thermal anomaly. Modern infrared receivers have a temperature coefficient of resistance, reaching tens of percent at 1°C. The typical thermal anomalies in the temperature contrast is $\Delta T = 10$°C. Energy contrast reaches values of 0.07–0.09, if the pixel size does not exceed the size of the thermal anomaly, and decreases linearly with the increase of the former. Minimum resolvable contrast to existing energy equipment is 0.4% and corresponds to the range of variation ratio of the characteristic size of thermal anomaly and a pixel in the range from 0.03 to 0.3, i.e. spatial resolution can substantially exceed the size of the thermal anomaly.

In addition to the spatial resolution, an important quantity for assessing the quality of the thermal and optical system is the probability of detecting thermal radiation from the object depending on signal/noise ratio, where the noise means temperature threshold of detection or, alternatively, temperature noise equivalent. The noise value is defined experimentally for specific optical systems and receives the equations of heat background light and varies from 0.1 to 0.2°C.

The method described provides a consistent and effective way if implementing operation related to decoding of information from the thermal anomaly as well as identifying its location and parameters. In this case, we consider the temperature field of the study area, mapped character images obtained under various shooting conditions, and analyze their dynamics with regard to the influence of all other factors. If the interpretation of thermal images is performed in conjunction with the data from visible or multispectral photography, it will facilitate recognition of terrain objects and allow the exclusion of anomalies of topographical nature, e.g. sun-warmed rock outcrops.

27.6 Sources of further information

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The modern methods of thermal photography and detection of hot objects are set out.

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The theory of formation of a boiler cavity and cleavage phenomena for underground nuclear explosions are set out.

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