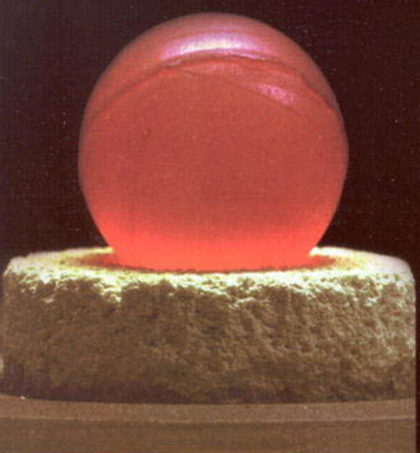


IMPROVING THE SCIENTIFIC BASIS FOR MANAGING DOE'S EXCESS NUCLEAR MATERIALS AND SPENT NUCLEAR FUEL

NATIONAL RESEARCH COUNCIL
OF THE NATIONAL ACADEMIES

IMPROVING THE SCIENTIFIC BASIS FOR MANAGING DOE'S EXCESS NUCLEAR MATERIALS AND SPENT NUCLEAR FUEL

Committee on Improving the Scientific Basis for Managing Nuclear Materials and Spent Nuclear Fuel through the Environmental Management Science Program

Board on Radioactive Waste Management

Division on Earth and Life Studies

NATIONAL RESEARCH COUNCIL
OF THE NATIONAL ACADEMIES

THE NATIONAL ACADEMIES PRESS
Washington, D.C.
www.nap.edu

THE NATIONAL ACADEMIES PRESS • 500 Fifth Street, N.W. • Washington, DC 20001

Support for this study was provided by the U.S. Department of Energy under Grant No. DE-FC01-99EW59049. All opinions, findings, conclusions, or recommendations expressed herein are those of the authors and do not necessarily reflect the views of the organizations or agencies that provided support for the project.

International Standard Book Number 0-309-08722-8 (book)
International Standard Book Number 0-309-50613-1 (PDF)

Additional copies of this report are available from:

The National Academies Press
500 Fifth Street, N.W.
Lockbox 285
Washington, DC 20055
(800) 624-6242 or
(202) 334-3313 (in the Washington Metropolitan Area);
Internet, http://www.nap.edu

COVER PHOTOS. Clockwise from top: Plutonium-238 from the Savannah River Site, South Carolina; Cesium-137 and Strontium-90 capsules at the Hanford Site, Washington; 14-ton cylinder containing depleted uranium hexafluoride at the Oak Ridge Reservation, Tennessee.

Printed in the United States of America

THE NATIONAL ACADEMIES

Advisers to the Nation on Science, Engineering, and Medicine

The **National Academy of Sciences** is a private, nonprofit, self-perpetuating society of distinguished scholars engaged in scientific and engineering research, dedicated to the furtherance of science and technology and to their use for the general welfare. Upon the authority of the charter granted to it by the Congress in 1863, the Academy has a mandate that requires it to advise the federal government on scientific and technical matters. Dr. Bruce M. Alberts is president of the National Academy of Sciences.

The **National Academy of Engineering** was established in 1964, under the charter of the National Academy of Sciences, as a parallel organization of outstanding engineers. It is autonomous in its administration and in the selection of its members, sharing with the National Academy of Sciences the responsibility for advising the federal government. The National Academy of Engineering also sponsors engineering programs aimed at meeting national needs, encourages education and research, and recognizes the superior achievements of engineers. Dr. Wm. A. Wulf is president of the National Academy of Engineering.

The **Institute of Medicine** was established in 1970 by the National Academy of Sciences to secure the services of eminent members of appropriate professions in the examination of policy matters pertaining to the health of the public. The Institute acts under the responsibility given to the National Academy of Sciences by its congressional charter to be an adviser to the federal government and, upon its own initiative, to identify issues of medical care, research, and education. Dr. Harvey V. Fineberg is president of the Institute of Medicine.

The **National Research Council** was organized by the National Academy of Sciences in 1916 to associate the broad community of science and technology with the Academy's purposes of furthering knowledge and advising the federal government. Functioning in accordance with general policies determined by the Academy, the Council has become the principal operating agency of both the National Academy of Sciences and the National Academy of Engineering in providing services to the government, the public, and the scientific and engineering communities. The Council is administered jointly by both Academies and the Institute of Medicine. Dr. Bruce M. Alberts and Dr. Wm. A. Wulf are chair and vice chair, respectively, of the National Research Council

www.national-academies.org

COMMITTEE ON IMPROVING THE SCIENTIFIC BASIS FOR MANAGING NUCLEAR MATERIALS AND SPENT NUCLEAR FUEL THROUGH THE ENVIRONMENTAL MANAGEMENT SCIENCE PROGRAM

WM. HOWARD ARNOLD, Chair, Westinghouse Electric Corporation (retired), Coronado, California
GREGORY B. COTTEN, U.S. Naval Academy, Annapolis, Maryland
KATHRYN A. HIGLEY, Oregon State University, Corvallis
LINN W. HOBBS, Massachusetts Institute of Technology, Cambridge
DONALD A. ORTH, E.I. du Pont de Nemours and Company (retired), Aiken, South Carolina
IRVIN OSBORNE-LEE, Prairie View A&M University, Texas
MARK T. PAFFETT, Los Alamos National Laboratory, New Mexico
DALE L. PERRY, Lawrence Berkeley National Laboratory, California
PER F. PETERSON, University of California, Berkeley
STEVEN M. THORNBERG, Sandia National Laboratories, Albuquerque, New Mexico
ROBERT W. YOUNGBLOOD, ISL, Inc., Rockville, Maryland

Board on Radioactive Waste Management Liaison

RODNEY C. EWING, University of Michigan, Ann Arbor

Staff

JOHN R. WILEY, Study Director
DARLA J. THOMPSON, Research Assistant
LAURA D. LLANOS, Senior Project Assistant

BOARD ON RADIOACTIVE WASTE MANAGEMENT

JOHN F. AHEARNE, Chair, Sigma Xi and Duke University, Research Triangle Park, North Carolina
CHARLES MCCOMBIE, Vice Chair, Consultant, Gipf-Oberfrick, Switzerland
ROBERT M. BERNERO, U.S. Nuclear Regulatory Commission (retired), Gaithersburg, Maryland
GREGORY R. CHOPPIN, Florida State University, Tallahassee
RODNEY EWING, University of Michigan, Ann Arbor
HOWARD C. KUNREUTHER, University of Pennsylvania, Philadelphia
NIKOLAY LAVEROV, Russian Academy of Sciences, Moscow
MILTON LEVENSON, Bechtel International (retired), Menlo Park, California
JANE C.S. LONG, Mackay School of Mines, University of Nevada, Reno
ALEXANDER MACLACHLAN, E.I. du Pont de Nemours and Company (retired), Wilmington, Delaware
NORINE E. NOONAN, College of Charleston, South Carolina
EUGENE A. ROSA, Washington State University, Pullman
ATSUYUKI SUZUKI, University of Tokyo, Japan
VICTORIA J. TSCHINKEL, The Nature Conservancy, Altamonte Springs, Florida

STAFF

KEVIN D. CROWLEY, Director
MICAH D. LOWENTHAL, Staff Officer
BARBARA PASTINA, Senior Staff Officer
JOHN R. WILEY, Senior Staff Officer
TONI GREENLEAF, Administrative Associate
DARLA J. THOMPSON, Research Assistant
LATRICIA C. BAILEY, Senior Project Assistant
LAURA D. LLANOS, Senior Project Assistant
ANGELA R. TAYLOR, Senior Project Assistant
JAMES YATES, JR., Office Assistant

Preface

The production of nuclear materials for the national defense was an intense, nationwide effort that began with the Manhattan Project and continued throughout the Cold War. Now many of these product materials, by-products, and precursors, such as irradiated nuclear fuels and targets, have been declared as excess by the Department of Energy (DOE). Most of this excess inventory has been, or will be, turned over to DOE's Office of Environmental Management (EM), which is responsible for cleaning up the former production sites. Recognizing the scientific and technical challenges facing EM, Congress in 1995 established the EM Science Program (EMSP) to develop and fund directed, long-term research that could substantially enhance the knowledge base available for new cleanup technologies and decision making.

The EMSP has previously asked the National Academies' National Research Council for advice for developing research agendas in subsurface contamination, facility deactivation and decommissioning, high-level waste, and mixed and transuranic waste. For this study the committee was tasked to provide recommendations for a research agenda to improve the scientific basis for DOE's management of its high-cost, high-volume, or high-risk excess nuclear materials and spent nuclear fuels. To address its task, the committee focused its attention on DOE's excess plutonium-239, spent nuclear fuels, cesium-137 and strontium-90 capsules, depleted uranium, and higher actinide isotopes.

The nuclear materials dealt with in this report are in relatively pure and concentrated forms, in contrast with waste and contaminated media dealt with in previous reports—in which radionuclides are typically dispersed at low concentrations in heterogeneous matrices. The committee concluded that not all of the excess nuclear materials are necessarily wastes; they cannot be re-created in the quantities now available, at least not without another effort approaching the Manhattan Project in scale, and some may have beneficial future uses. Research funded by the EMSP and other organizations should be directed primarily at discovering such uses, safely stabilizing the inventory, and developing a scientific basis for future disposition options.

In conducting this study, the committee held six meetings and visited four DOE sites. We recognize that a great deal of effort went into arranging presentations to the committee by DOE and contractor personnel. We especially thank Mark Gilbertson and Ker-Chi Chang of DOE headquarters for their help throughout the study. Our visit coordinators at the sites were Allen Croff, Oak Ridge National Laboratory; Jay Bilyeu, DOE-Savannah River; Alan Riechman, Savannah River Technology Center; and Marcus Glasper, DOE-Richland. Committee members Mark Paffett, Los Alamos National Laboratory (LANL), and Steven Thornberg, Sandia National Laboratories (SNL) also arranged, respectively, the visit to LANL and discussions with SNL scientists in Albuquerque, New Mexico.

We also recognize the staff of the National Academies' Board on Radioactive Waste Management (BRWM) for their assistance during the study. John Wiley, who served as study director, helped to guide the committee through its fact finding, report writing, and report review. Rodney Ewing, BRWM liaison, provided much helpful advice. Staff members Laura Llanos and Toni Greenleaf were always efficient and cheerful as they handled all of the many logistic details for the committee.

Finally, I want to thank the members of the committee. They were a pleasure to work with, and each made significant contributions.

Wm. Howard Arnold
Chairman

List of Report Reviewers

This report has been reviewed in draft form by individuals chosen for their diverse perspectives and technical expertise, in accordance with procedures approved by the National Research Council (NRC) Report Review Committee. The purpose of this independent review is to provide candid and critical comments that will assist the institution in making the published report as sound as possible and to ensure that the report meets institutional standards for objectivity, evidence, and responsiveness to the study charge. The content of the review comments and draft manuscript remains confidential to protect the integrity of the deliberative process. We wish to thank the following individuals for their participation in the review of this report:

Cynthia Atkins-Duffin, Lawrence Livermore National Laboratory
Harold Beck, U. S. Department of Energy Environmental Measurements Laboratory (retired)
David Clark, Virginia Tech
Norman Eisenberg, University of Maryland
Charles Forsberg, Oak Ridge National Laboratory
Milton Levenson, Bechtel International (retired)
Alexander MacLaughlin, E.I. du Pont de Nemours & Company (retired)

Although the reviewers listed above have provided many constructive comments and suggestions, they were not asked to endorse the conclusions or recommendations, nor did they see the final draft of the report before its release. The review of this report was overseen by Chris G. Whipple, ENVIRON International Corporation. Appointed by the National Research Council, he was responsible for making certain that an independent examination of this report was carried out in accordance with NRC procedures and that all review comments were carefully considered. Responsibility for the final content of this report rests entirely with the authoring committee and the NRC.

Contents

APPENDIXES

Executive Summary

Nuclear weapons production in the United States was a complex series of integrated activities carried out at 16 major sites and over 100 smaller ones. Production stopped abruptly in 1992 at the end of the Cold War leaving a legacy of radioactive wastes, contaminated media and buildings, and surplus nuclear materials. Focusing on the last of these categories, the statement of task for this report directed the committee[1] to provide recommendations on a research agenda that would improve the scientific basis for the Department of Energy's (DOE's) management of its inventory of high-volume, high-cost, or high-risk spent fuel and nuclear materials. To this end the committee focused its attention on the following:

- Plutonium-239. About 50 metric tons of this isotope, a principal component in nuclear weapons, have been declared excess. DOE intends to convert most excess Pu-239 into mixed oxide fuel for use in commercial reactors. About 17 metric tons of the excess are in the form of impure scraps and residues for which conversion may be difficult.
- Spent nuclear fuel. DOE manages a wide variety of fuel types, which total approximately 2,500 metric tons. Many fuels are corroding, and their processing or disposal is many years away.
- Cesium-137 and strontium-90 capsules. Approximately 2,000 capsules stored at the Hanford, Washington, site contain a total of 67 million curies[2] of radioactivity within a volume of only

---

[1] The Committee on Improving the Scientific Basis for Managing Nuclear Materials and Spent Nuclear Fuel Through the Environmental Management Science Program is referred to as "the committee" throughout this report.

[2] DOE literature typically expresses radioactivity in units of curies rather than becquerels.

about 5 cubic meters. These capsules represent almost 40 percent of the radioactivity at the Hanford site and have been described as the most lethal source of radiation in the United States, except for the core of an operating nuclear reactor.

- Depleted uranium. A residue from uranium enrichment operations, DOE's inventory includes over 700,000 metric tons of uranium hexafluoride (UF_6), which can produce toxic gases by reacting with moisture and air. Most is stored at three sites in 14-ton carbon steel canisters, many of which are badly corroded, and some have leaked. DOE intends to convert the UF_6 to a more stable oxide. Disposition[3] plans for the oxide have not yet been determined.
- Higher actinides. Including neptunium-237, americium-243, and curium-244, these are materials that can no longer be produced in the United States in the kilogram quantities now available. Continued storage is expensive and presents potential health risks; discarding them may prove to be an irrevocable loss of a unique asset.

Cleaning up the Cold War legacy is the mission of DOE's Office of Environmental Management (EM). In 1995, Congress chartered the Environmental Management Science Program (EMSP) to bring the nation's scientific capability to bear on the difficult, long-term cleanup challenges facing DOE. To fulfill its charter, the EMSP solicits proposals and selectively funds research on problems relevant to the needs of EM. This report completes the fifth in a series of studies requested by the EMSP to assist in developing its calls for proposals and evaluating proposals. The previous studies (NRC, 2000, 2001a, 2001b, 2002) dealt with waste and site cleanup. A significant difference with the excess nuclear materials dealt with in this report is that most have not been declared as waste. The statement of task for this study accordingly directed the committee to identify research opportunities for storage, recycle, or reuse as well as disposal of these materials.

Findings and Recommendations

The overarching theme throughout this study is that scientific research beginning now can inform DOE's future decisions for permanent disposition of surplus nuclear materials. A salient characteristic of

---

[3] Throughout this report, the term "disposition" includes options such as storage, reuse, and disposal.

nuclear materials is their potential for unforeseen, beneficial future uses. DOE should avoid decisions today that foreclose future options.

The EMSP should emphasize research toward stabilizing DOE's excess nuclear materials and discovering beneficial uses for these materials.

There is a tension between the needs of today's milestone-driven decisions and the planning of longer-term research. Meeting programmatic milestones is a primary objective for EM. Research priorities have been tied to these milestones. Such a narrow focus may foreclose research that can lead to fundamentally new concepts and opportunities.

The committee was guided in its deliberations by considering a different role for research, namely, preparing to make more informed programmatic decisions in the future. This is a better approach than trying to settle all decision making now, for all time, in light of substantial uncertainties (see also NRC, 2003). This approach implies a program of research that is not restricted by current milestones or assumptions about future needs.

The nuclear materials dealt with in this report have been available for only a few decades. Basic physical and chemical principles guarantee that there will be no simple, shortcut ways to replace the currently available quantities of nuclear materials that resulted from 50 years of intense effort in the United States' massive nuclear complex. The next few decades may bring unforeseen beneficial uses so that these materials are recognized as valuable and irreplaceable resources.

Plutonium-239

Making the plutonium isotope of mass 239 (Pu-239) was a principal objective of nuclear materials production in the United States from the 1940s through the late 1980s. Approximately 100 metric tons of Pu-239 were obtained from the nuclear reactors and separations facilities at the Hanford, Washington, and the Savannah River, South Carolina, sites for use in nuclear weapons (see Chapter 3 and Appendix A). According to current U.S. policy, about half of this product has been declared as surplus. The surplus inventory includes clean metal—mainly from disassembly of weapons—oxide, and plutonium combined with a variety of other materials in reactor fuels, targets, and miscellaneous forms.

DOE's disposition options for surplus Pu-239 include:

- storage according to the DOE 3013 Standard for up to 50 years;
- fabrication into mixed oxide (MOX) fuel;
- disposal as transuranic (TRU) waste in the Waste Isolation Pilot Plant (WIPP); and
- disposal along with high-level waste and spent fuels, e.g., in the planned Yucca Mountain, Nevada repository.

A key element in DOE's strategy for eventual disposal of its inventory is the conversion of as much of the excess Pu-239 as is technically and economically feasible into MOX fuel for commercial power reactors.[4] The spent MOX fuel would be co-disposed with other spent nuclear fuels. However, approximately 17 metric tons of excess Pu-239 are in the form of scraps and residues, including very impure materials. The disposition of this material is uncertain and will present technical challenges for MOX operations.

The EMSP should support research to help maximize the portion of DOE's excess Pu-239 inventory that can be used as MOX fuel and that will support the scientific basis for disposal of impure plutonium not suitable for MOX fuel. Research should include fundamental chemistries for storing and purifying plutonium, modeling of MOX fuel performance to help ensure reactor safety, and devising high-integrity, theft-resistant forms for disposal.

Research opportunities for storage include study of long-term corrosion and gas generation in the sealed 3013 canisters (see Chapter 3), process analytical chemistry and materials characterization for MOX fabrication, and improved moisture analysis and nondestructive assay techniques for use in high-radiation environments. For less pure materials that may not be directly suitable for MOX fabrication, research is needed to improve the characterization and separation of undesirable impurities to make more material available for MOX and potentially to allow greater flexibility in incorporation of a wider range of materials into MOX than current specifications allow.

The committee believes there will likely be impure Pu-239 materials that cannot be converted to MOX, but nevertheless are too rich for disposal as TRU waste in the WIPP. Further research into alternate ways of immobilizing this material, for example, in ceramic matrices, to meet criteria for co-disposal with high-level waste and spent fuel is needed. In addition, there are potential crosscutting research topics on stabilization of spent fuel and plutonium residues for storage and disposal.

---

[4] The committee did not review the MOX fuel program.

Spent DOE Nuclear Fuel

DOE manages an assortment of over 250 spent nuclear fuel (SNF) types that altogether comprise about 2,500 metric tons of heavy metal (MTHM).[5] DOE spent fuel was generated in military and civilian reactor development, research, and fuel testing programs. The inventory also includes irradiated fuel and target[6] assemblies that were placed in storage when DOE stopped reprocessing nuclear fuel for production purposes in 1992. DOE plans to dispose of its SNF along with commercial SNF and vitrified high-level waste in a repository at Yucca Mountain. Because DOE has only recently begun to prepare a license application for Yucca Mountain, uncertainty exists in the future waste acceptance criteria for the various types of DOE spent fuel.

Most types of DOE spent fuel have important characteristics that are different from commercial spent fuel, which will comprise most of the waste disposed in Yucca Mountain, if licensed and constructed. These are primarily differences in the chemical forms of the fuel and the cladding materials that encase it, and the isotopic composition of the fuel. The different characteristics affect the spent fuel's chemical stability and potential for gas generation, decay heat generation and potential for thermal damage under different storage and accident conditions, potential for inadvertent nuclear criticality, and attractiveness of the material for theft.

The EMSP should support research to help ensure safe and secure storage and disposal of DOE SNF. Research should emphasize materials characterization and stabilization, including developing a better understanding of corrosion, radiolytic effects, and accumulated stresses. This research should be directed toward determining a limited number of basic parameters that can be used to evaluate the long-term stability of each of the types of DOE SNF in realistic storage or repository environments.

The primary research challenge and opportunity in characterization is nondestructive assay of plutonium and other isotopes in the high-radiation environment that is typical of most spent fuels. Interim storage

---

[5] MTHM refers to the mass of uranium and/or plutonium used to fabricate the fuel. It does not include the mass of the fuel cladding or ancillary components.

[6] Most of DOE's nuclear materials were created in nuclear reactors through the capture of neutrons by various target isotopes, e.g., U-238 (see Appendix A). Using separate fuel (driver) and target assemblies increased production efficiency. DOE manages irradiated targets as SNF. The committee does not distinguish between fuels and targets when referring to SNF.

requires conditioning methods that are inexpensive but provide sufficient stability to meet safety requirements for several decades. For spent fuels of relatively low chemical stability, such as DOE aluminum-clad spent fuels, a wide variety of potential degradation mechanisms exist: radiolytic gas generation, biocorrosion, pitting corrosion, interactions with other materials in storage containers, oxidation, matrix dissolution, and hydriding. Stresses can accumulate from the fuel's thermal history and from other effects such as swelling due to oxidation or radiolytic displacements and transmutations. There are opportunities for research to better understand these degradation mechanisms and to identify inexpensive approaches to arrest them.

Because disposal criteria are uncertain, research is needed to provide bases for a variety of conditioning methods. Minimal conditioning may prove to be problematic for highly enriched uranium fuels, due to criticality issues, and for aluminum-clad fuels, due to chemical stability issues. Research to further develop reprocessing options where the spent fuel is dissolved in a molten salt or an aqueous solution and separate streams of well-characterized materials are created may help to address the specific issues of high enrichment and cladding stability. There are opportunities for collaboration with the new DOE Advanced Fuel Cycle Initiative to identify research that would make the reprocessing approach viable for some DOE spent fuels that would otherwise have difficulty meeting repository waste acceptance criteria.

Cesium-137 and Strontium-90 Capsules

In the early 1970s operators at the Hanford site removed a large fraction of the Cs-137 and Sr-90 from the site's high-level tank waste in order to reduce the requirements for cooling the tanks. The cesium and strontium were concentrated and sealed in stainless steel capsules for potential uses, for example, thermoelectric generators or sterilizers. The expected applications for the Hanford capsules did not materialize, and ceased entirely in 1988 after a capsule being used in the commercial sector was found to be leaking. The almost 2,000 capsules, stored underwater at the Waste Encapsulation and Storage Facility (WESF), contain a total of 67 million curies of radioactivity—approximately 37 percent of the total radioactivity at the Hanford site (see cover photograph). The disposition of these capsules has not been decided; options include:

- continued underwater storage at the WESF facility,
- passive storage in air at a new facility,
- overpacking and disposal of the capsules in a geologic repository, and

- incorporating the isotopes into a glass or crystalline matrix for disposal in a geologic repository.

The EMSP should support research that will help ensure continued safe storage and potential use or eventual disposal of the Hanford Cs-137 and Sr-90 capsules. Research should lead to understanding potential failure mechanisms of the present capsules, ways to convert the isotopes to stable glass or ceramic forms, and understanding long-term hazards of disposition options.

There are opportunities for fundamental research to understand the chemical and physical alterations of CsCl and SrF_2 under intense radiation, localized heating, and change of valence states accompanying radioactive decay. CsCl and SrF_2 are susceptible to partial radiolytic decomposition to colloidal metal particles and evolvable halogen gas in the temperature range 100–200 °C after accumulated ionization doses in the dose region 10^8–10^{10} Gy. Cesium-137 (monovalent) decays into barium-137 (divalent), and strontium-90 (divalent) decays into zirconium-90 (normally tetravalent) via a short-lived yttrium-90 intermediate. These transmutations lead to very different physical and chemical properties, such as melting and phase-transition points, bulk volume changes, and changes in the ionic radii. Ionization due to the intense radiation fields is likely to induce other changes.

Capsule integrity is essential for interim storage. Twenty-three cesium capsules have been placed in overpacks because they have swollen or otherwise been damaged. Reasons for the swelling are not well understood. There are opportunities for research toward understanding the possible failure mechanisms and predicting incipient failures.

Because the materials in the capsules are concentrated and relatively pure, they are good candidates for incorporation into crystalline matrices that could be developed to be robust against heat, radiation, and transmutations. For vitrification, research is needed to ensure that the isotopes can be sufficiently dispersed in a glass matrix to avoid detrimental effects of heat and radiation in long-term storage or disposal.

Depleted Uranium

Most depleted uranium (DU) is in the chemical form of uranium hexafluoride (DUF_6) amounting to 450,000, 198,000, and 56,000 metric tons, stored at DOE sites near Paducah, Kentucky; Portsmouth, Ohio; and Oak Ridge, Tennessee, respectively. The DUF_6 is stored in cylinders stacked in open-air storage yards. Each contains about 14 tons of DUF_6 (see cover photograph). The Oak Ridge Reservation has the oldest of these cylinders, some dating back to the Manhattan Project. The most immediate risk posed by the DUF_6 is its potential to react with moisture to form hydrogen fluoride, a highly corrosive and chemically toxic gas.

DOE has recently taken a first step toward dispositioning its DUF_6 by awarding an 8-year contract to Uranium Disposition Services to build and operate facilities at Paducah and at Portsmouth to convert it to the stable oxide U_3O_8. The Portsmouth plant will also convert the Oak Ridge DUF_6. The contractor will store the oxide at the two conversion facilities. Options for future disposition of the DU, once converted to oxide, are continued storage, reuse, or disposal as waste. Recent concerns over the health effects of DU have led to a resurgence of research on its health effects, but significant gaps remain. Beneficial ways to reuse large amounts of uranium have not been identified.

The EMSP should support near-term (1–5-year) research to help ensure safety of the DUF_6 during storage, transportation, and conversion. The EMSP should also support longer-term research that might lead to new, beneficial uses for uranium or that would provide a scientific basis for selecting a disposal option.

The way the cylinders are stacked in the storage yards restricts the workspace between cylinders and in some cases precludes workers from being able to examine the entire outer surface of each cylinder. Nor is it possible to confidently move and hoist all cylinders because corrosion may have weakened some to the point that they could be damaged by the available handling techniques and equipment—a problem that will increase as time passes. There is need and opportunity for near-term research that will support DOE's plans for converting its DUF_6 to oxide. For example, robotic or remotely operated methods to assess the integrity of the cylinders, extract DUF_6 from those that cannot be moved safely, and measure radioactive contaminants (some contain low levels of fission products from recycled uranium) would enhance worker safety.

Research to exploit the special chemical and metallurgical properties of uranium for new uses could convert this large amount of material from a disposal problem to an asset. There are opportunities to use recent advances in biology to develop a better understanding of the potential health effects of uranium metal, oxide, and typical compounds. This research can help establish a scientific basis both for new uses of DU or for its eventual disposal. For disposal, research to develop a scientific basis for returning the material to a former uranium mine or mined cavity is recommended.

The Higher Actinides

With the closure of its production reactors and separations facilities, DOE no longer has the capability for large-scale production of higher actinide isotopes,[7] most of which were made in special campaigns that involved multiple irradiation and separation steps (see Chapter 7 and Appendix A). Currently there is little or no use foreseen for the kilogram quantities of these isotopes that are in storage, and for the most part they are considered a liability by EM. The facilities for handling and storing these isotopes are being closed as part of site cleanup. Consequently, EM plans to dispose of many unique materials as waste, e.g., by mixing with high-level tank waste. This route would foreclose all other options and risks future regret of an irrevocable action.

The EMSP should support research to preserve and stabilize the inventory of higher actinide isotopes, identify beneficial new uses, and develop a better understanding of their radiological and chemical health effects.

The higher actinides in the DOE inventory represent material that may be useful in its present form, may be suitable for target material, or may be essential for research into developing new materials. The committee concluded that there are three principal challenges to preserving the inventory:

- Facilities capable of handling or storing the materials are being closed.
- Few new nuclear scientists are being trained.
- Accumulated knowledge, both documentation and personal expertise, is being lost.

The Office of Science has an opportunity to lead other DOE offices and industrial partners in establishing a center of excellence to ensure that the United States has a continuing capability to handle and store large inventories of higher actinides for research, beneficial use, or as feedstock. EMSP-funded research directed at both fundamental science and new uses of the higher actinides can be an important step toward preserving the inventory.

---

[7] Defined for the purpose of this report as isotopes having an atomic number of 93 (Np) or greater.

Research Priorities

The EMSP's congressional charter calls for long-term, path-breaking research. In addition, opportunities for research that provides a high potential payoff in addressing urgent, near-term needs may arise. As a practical matter, the EMSP may well encounter a range of research opportunities that span short- and long-term needs.

Opportunities for research that might provide shorter-term (1-5 year) payoffs are generally in the area of stabilizing the inventory for storage. Specific examples include stabilizing Pu-239 for 50 years of storage according to the DOE 3013 Standard, arresting the cladding degradation on some DOE spent fuels and preparing them for decades of storage before eventual disposal, and supporting DOE's plans to convert its DUF_6 to a stable oxide.

Begun now, longer-term research would feed a continuously growing body of scientific information to support decision making and have the potential of providing scientific breakthroughs. Longer-term research should be directed toward beneficial new uses for DOE's nuclear materials or their disposal.

This report is the last in a series of five National Academies' studies requested by the EMSP to assist in providing an agenda for research to support and enhance DOE's site cleanup program. The previous reports dealt exclusively with environmental contamination and waste issues. Most of the excess nuclear materials that are the subject of this report have not been declared as waste, and according to its statement of task the committee emphasized research directed toward preserving and reusing the materials.

Nevertheless, there is a broad consistency among the recommendations in all five studies. Three areas stand out as offering opportunities for the EMSP to support scientific research that crosscuts most of DOE's cleanup challenges:

- characterization of fundamental chemical and physical, and biological properties of the materials, wastes, or contaminated media;
- treatment to ensure near- and long-term stability, including understanding the fundamental parameters that affect stability; and
- assessment of health or environmental risks.

By focusing its limited funds in these crosscutting areas and by leveraging funding by cooperative research with other DOE offices or the private sector, the EMSP is most likely to achieve the scientific breakthroughs intended by its congressional charter.

1

Introduction, Background, and Task

The Department of Energy's (DOE's) Environmental Management Science Program (EMSP) was established by the 104th Congress[1] to bring the nation's basic science infrastructure to bear on the massive environmental cleanup effort under way in the DOE complex. The objective of the EMSP is to develop and fund a targeted, long-term research program that will result in transformational or breakthrough approaches for solving the department's environmental problems. The goal (DOE, 2000a, pp. 1-2) is to support research that will

- Lead to significantly lower cleanup costs and reduced risks to workers, the public, and the environment over the long term.
- Bridge the gap between broad fundamental research that has wide-ranging applicability . . . and needs-driven applied technology.
- Serve as a stimulus for focusing the nation's science infrastructure on critical national environmental management problems.

To help meet these goals, the EMSP provides 3-year competitive awards to investigators in industry, national laboratories, and universities to undertake research on problems relevant to DOE cleanup efforts. From its inception in 1996, the EMSP has provided $327 million in funding for 399 research projects.

This study, addressing DOE's excess nuclear materials and spent nuclear fuels, is the fifth study undertaken by the National Academies' National Research Council (NRC) to assist DOE in developing a research agenda for the EMSP.[2] The previous four reports (NRC, 2000, 2001a, 2001b, 2002) gave advice for research in subsurface contamination,

---

[1] Public Law 104-46, 1995.

[2] An initial study advised DOE on establishing the EMSP (NRC, 1997a).

high-level waste, facility deactivation and decontamination, and transuranic and mixed wastes. DOE has used these studies in developing calls for research proposals and for evaluating proposals submitted.

After its establishment by Congress and through most of the course of this study, the EMSP was managed by a partnership between the DOE Office of Environmental Management (EM), which has primary responsibility for the cleanup mission, and the DOE Office of Science, which manages basic research programs. The advice provided by the NRC studies, as well as the EMSP's calls for proposals, reflected EM's organization of its science and technology development activities into five "focus areas," which are the topical areas of the NRC studies mentioned above—subsurface contamination, high-level waste, facility deactivation and decommissioning, transuranic and mixed wastes, and nuclear materials.

During the course of this study the EMSP was in transition from EM to the DOE Office of Science, Environmental Remediation Sciences Division (ERSD).[3] The committee did not attempt to assess or comment on this transition, but rather focused its attention on research needs and opportunities for DOE's excess nuclear materials. However, the committee joins the four previous NRC committees in noting that the program's approximately $30 million annual budget allocation can support only a small fraction of the research agenda that is needed to address the EMSP's objectives.

With its new home in the Office of Science, the EMSP has a better opportunity to coordinate its research programs with other DOE offices (e.g., Office of Civilian Radioactive Waste Management, Office of Nuclear Energy, Science and Technology) and non-DOE organizations (e.g., Department of Defense, National Science Foundation) that are supporting similar research. Such cooperation might be along disciplinary rather than programmatic lines. Maintaining relevance to EM problems may be more difficult after the transition. Since July 2000, ERSD has been developing a strategic plan to better integrate the EMSP with other environmental research and development programs (Patrinos, 2002). The plan had not been finalized at the time this report was completed.

Statement of Task

The statement of task for this study charged the committee to provide recommendations for a science research program for treatment, storage, and recycle, reuse, or disposal[4] of nuclear materials, including

---

[3] See http://www.sc.doe.gov/.

[4] Throughout this report, the term "disposition" is used to denote options such as storage, reuse, recycle, or disposal.

SIDEBAR 1.1 STATEMENT OF TASK

The objective of this study is to provide recommendations to the Environmental Management Science Program on a research agenda to improve the scientific basis for treatment, storage, and recycle/reuse/disposal of spent nuclear fuel and nuclear materials (including depleted uranium) that are currently being managed by DOE or will be generated in the future as part of DOE's cleanup program. The study will accomplish the following:

- **Evaluate the current state of treatment, storage, and "end state" (i.e., disposal, recycle, or reuse) options for high-volume, high-cost, and high-risk spent fuel and nuclear materials managed by DOE.**
- **Identify gaps in the scientific basis for treating, storing, recycling, reusing, or disposing of these materials.**
- **Identify a research agenda to address these gaps and add to scientific knowledge generally, taking into account the levels of funding likely to be available to the program in future years as well as research funded by other programs. This agenda should provide, if possible, an estimate of the time that will be required to obtain the necessary scientific bases for advances.**

depleted uranium and spent nuclear fuels, that are currently being managed by DOE or will be produced as part of DOE's site cleanup program (see Sidebar 1.1).

To address the statement of task, the committee directed its attention to the following five groups of materials that it believes present the most difficult challenges and provide the greatest opportunities for EMSP research to significantly improve DOE's ability to manage its excess nuclear materials and spent nuclear fuels:

- Plutonium-239. The inventory of Pu-bearing solids that is considered DOE legacy material encompasses approximately 100 metric tons, about half of which has been declared as excess. Much of the excess includes scraps and residues that have uncertain disposition routes. The material presents significant technical challenges, and potential health and security risks.
- Spent nuclear fuel. DOE manages a wide variety of fuel types, which total approximately 2,500 metric tons. Many fuels are corroding, and their processing or disposal is many years away. They present significant technical challenges and potential health risks. Highly enriched uranium fuels may present security risks.
- Cesium-137 and strontium-90 capsules. Approximately 2,000 capsules stored at the Hanford, Washington, site contain a total of 67 million curies of radioactivity within a volume of only about 5 cubic meters. These capsules represent almost 40 per-

cent of the radioactivity at the Hanford site and have been described as the most intense single source of radiation in the United States, except inside the core of an operating nuclear reactor. Their disposition presents significant technical challenges and potential health risks.

- Depleted uranium. A residue from uranium enrichment operations, DOE's inventory includes over 700,000 metric tons of uranium hexafluoride (UF_6), which can produce toxic hydrogen fluoride and uranyl compounds upon reacting with moisture and air. Most is stored at three sites in 14-ton carbon steel canisters, many of which are badly corroded and some of which have leaked. DOE intends to convert the UF_6 to a more stable oxide. Disposition plans for the oxide have not yet been determined.
- Higher actinides. Including neptunium-237, americium-243, and curium-244, these are materials that can no longer be produced in the United States in the kilogram quantities that now exist. Continued storage is expensive and presents potential health risks; discarding them may prove to be an irrevocable loss of a unique asset.

Some excess nuclear materials in the DOE inventory (e.g., U-233, thorium) were not considered by the committee because they appear to present fewer challenges in terms of risk, volume, cost, or unique research opportunities as those in the five groups selected (see DOE, 2000c). Among those selected, the committee sought to be comprehensive in identifying scientifically valid and relevant research, realizing that only a fraction of this research can be funded. The general research recommendations were developed through a consensus process that considered input to the committee, site needs, the existence of critical knowledge gaps, the potential for future cost and schedule savings, and the possibility of achieving scientific breakthroughs.

Chapter 2 of this report describes the origin, disposition options, and challenges of nuclear materials in the DOE complex. The chapter also frames the committee's views on how new knowledge gained through EMSP research can assist DOE's broader, long-term decisions for managing and dispositioning its excess nuclear material.

Chapters 3 though 7 address each of the five sets of materials. In each chapter, an overview describes the current status and DOE's plans for dealing with the material. Technical gaps and challenges, as determined from the committee's fact-finding visits to DOE sites, literature reviews, and deliberations, are presented. The committee then gives its general recommendation and describes opportunities for EMSP research to address the gaps and challenges and to enhance scientific knowledge generally.

Chapter 8 summarizes the research recommendations and suggests a broad prioritization of near-term (1-5 year) and longer-term nuclear materials research. Because this report is the last in a series of five National Academies' reports that have suggested research agendas for the EMSP, the committee also provided a summary of research that crosscuts all five reports.

The committee held six meetings between October 2001 and September 2002 to gather information (see Appendix C). The committee's fact finding included site visits and briefings at the Los Alamos National Laboratory (New Mexico), Oak Ridge Reservation (Tennessee), Savannah River Site (South Carolina), and the Hanford Site and Pacific Northwest National Laboratory (Washington). The committee also received briefings by DOE headquarters personnel. Especially useful were roundtable discussions among the committee and scientists from Los Alamos and Sandia National Laboratories (New Mexico), Oak Ridge, Savannah River, Hanford, and Pacific Northwest National Laboratory.

2

The Challenges of Managing DOE's Excess Nuclear Materials

Nuclear weapons production in the United States was a complex series of integrated activities carried out at 16 major sites and over 100 smaller ones. Production stopped abruptly in 1992 at the end of the Cold War leaving a legacy of radioactive wastes, contaminated media and buildings, and surplus nuclear materials.[1] Site cleanup and closure is the mission of the Department of Energy's (DOE's) Office of Environmental Management (EM). Previous National Academies' studies have assisted the EM Science Program (EMSP) in developing a research agenda for waste and site cleanup (NRC, 2000, 2001a, 2001b, 2002). A significant difference with the excess nuclear materials dealt with in this report is that most have not been declared as waste, and disposition paths have not been decided. The statement of task for this study accordingly directed the committee to identify research opportunities for storage, recycle, or reuse as well as disposal of these materials. The surplus nuclear materials dealt with in this report differ from waste and contaminated media in several important ways:

- Most nuclear materials in the inventory are in concentrated, relatively pure forms.
- The United States can no longer produce these materials in quantities that approach those of the inventory.
- Some of the materials may have beneficial future uses.
- Some materials, for example, plutonium and spent nuclear fuels, present security concerns.

DOE's strategy for managing these materials is to collect them at a few of its larger sites (Hanford, Washington; Savannah River, South Car-

---

[1] Civilian nuclear energy research by DOE and its predecessors created additional nuclear materials that are now in DOE's inventory, as did naval propulsion activities.

olina; Oak Ridge, Tennessee; Idaho National Engineering and Environmental Laboratory) to allow "de-inventorying" and closing other sites. Consolidating the materials onto fewer sites has practical advantages, such as security and cost effectiveness, but the long-term character of the materials management problem remains.

As discussed later in this chapter, the committee concluded that the EMSP should foster research to reduce uncertainty in current plans for dispositioning its surplus nuclear materials and to improve the scientific basis for future decisions. Research emphasis should be on stabilizing the separated materials and developing beneficial uses. Because of its limited budget, the EMSP should coordinate its nuclear materials research with other programs in the Office of Science, EM, and the National Nuclear Security Administration.

DOE's Former Nuclear Materials Production

DOE's production era activities that led to its current inventory of nuclear materials can be summarized as follows:[2]

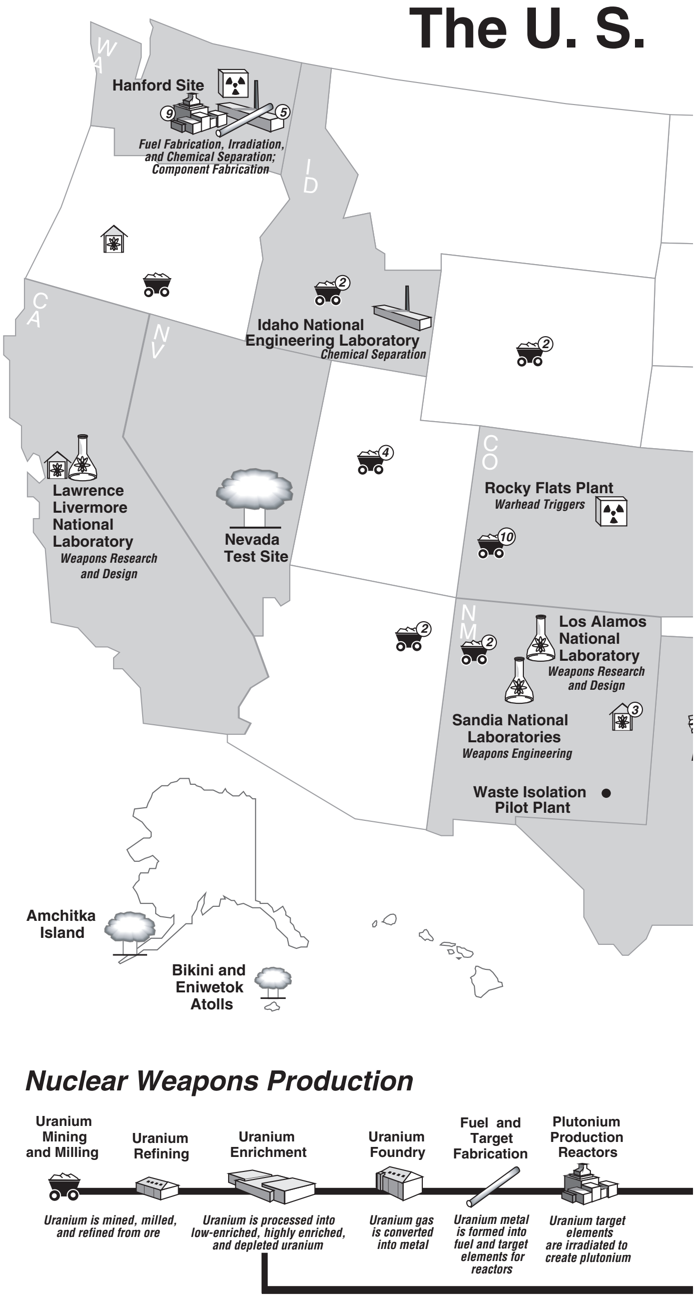
- uranium mining, milling, refining, and isotope enrichment;
- nuclear reactor fuel and target fabrication;
- reactor operations;
- chemical separations;
- weapon component fabrication;
- weapon assembly, maintenance, modification, and dismantlement.

The focus of DOE's work was making plutonium and tritium for nuclear weapons (see Figure 2.1). Approximately 100 metric tons of Pu-239 were obtained from the production reactors and separations facilities at the Hanford and the Savannah River sites. About half of this inventory has been declared as surplus. The surplus includes clean metal from weapon disassembly and other sources, and impure metals, oxides, and other forms such as scraps and residues that were in process or stored when production operations ceased. The committee concluded that managing plutonium presents the greatest excess nuclear material challenge for DOE and that research should help sup-

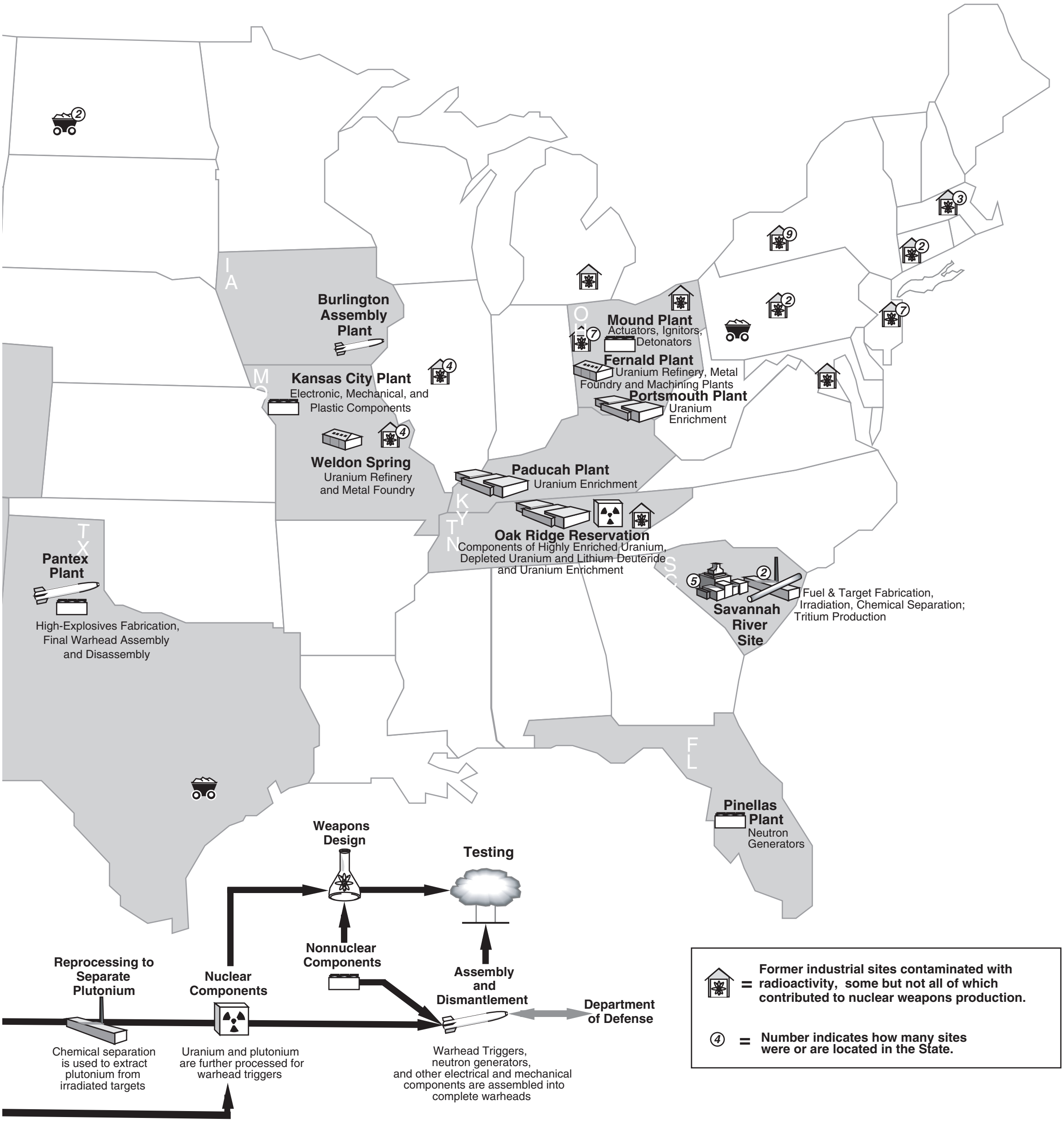
---

[2] See Appendix A for a more detailed description of nuclear materials production in the DOE complex.

Figure 2.1. The United States nuclear weapons complex included facilities that were constructed throughout the country. This figure indicates the location of some of the major facilities and depicts the key production steps. Source: DOE, 1996a.



Nuclear Weapons Complex



port DOE's plans for storing and beneficially reusing its Pu-239, as described in Chapter 3.

Reactor operations created the plutonium and essentially all other isotopes managed throughout the DOE complex. Enriched uranium served as fuel in production reactors, and excess neutrons from the nuclear chain reaction bred Pu-239 and other isotopes in "targets" made of depleted uranium. Irradiated spent fuel and targets were routinely reprocessed to recover the plutonium, uranium, and other isotopes. However, when the United States stopped its plutonium production, some 250 fuel types amounting to about 2,500 metric tons of heavy metal[3] of spent nuclear fuel and targets were left unreprocessed. Most are stored at Hanford, Idaho, Savannah River, and Oak Ridge. While DOE's spent nuclear fuel (SNF) inventory is only about 5 percent of the inventory of spent power reactor fuels managed by the commercial sector, DOE is challenged with a wide variety of fuel types—some of which are deteriorating. As described in Chapter 4, research should focus on means to ensure that these fuels are stabilized for several decades of storage and that they will meet yet to be defined acceptance criteria for disposal in a geological repository.

In addition to separating the desired products, reprocessing generated large volumes of highly radioactive waste, which were stored mainly in million gallon capacity tanks at the reprocessing sites. Most significant among the longer-lived, heat-producing fission products in the high-level waste are strontium-90 and cesium-137. In the early 1970s, Hanford removed a large fraction of these isotopes from its tank waste in order to reduce the heat produced in the tanks, and concentrated the isotopes in capsules for potential uses (thermoelectric generators, sterilizers). The almost 2,000 capsules contain about 67 million curies of radioactivity, approximately 37 percent of the total radioactivity at the Hanford Site. Their heat and intense radioactivity present challenges for their eventual disposition as well as research opportunities to support disposition plans, as described in Chapter 5.

Enriched uranium, used to fabricate reactor fuels and weapon components, resulted from multistep processes that gradually concentrated the fissile isotope U-235, which comprises only about 0.7 percent of natural uranium. Enriching a portion of the uranium in U-235 created a massive legacy of about half a million tons of uranium (metal equivalent) depleted in U-235. This depleted uranium is stored as uranium hexafluoride (UF_6) in large cylinders at the former enrichment sites near Paducah, Kentucky; Portsmouth, Ohio; and Oak Ridge, Tennessee.

---

[3] "Heavy metal" refers to the mass of uranium and/or plutonium in the fuel.

Chapter 6 describes research needs and opportunities for managing this very large amount of slightly radioactive, chemically toxic material.

DOE also used its production reactors and chemical separation facilities in a number of campaigns to produce isotopes for special applications (Pu-238 for thermoelectric power in space vehicles, see cover photograph; Cf-252 for cancer treatment). Most resulted from multiple irradiation and separation steps, which eventually built up the higher actinide isotopes through successive neutron captures. The shutdown of the DOE's production reactors and separations facilities precludes the future, large-scale manufacture of these isotopes. As DOE continues to close not only its production facilities, but also other facilities capable of handling and storing these isotopes, the potential benefits of these unique materials may be lost. Research needs and opportunities that may lead to future beneficial uses of these isotopes or aid DOE in deciding how to disposition these materials are discussed in Chapter 7.

Disposition Options

DOE has developed a comprehensive set of roadmaps for dispositioning essentially all of its nuclear materials (Tseng, 2001). In most instances there are multiple disposition options, but most eventually lead to disposal end points, for example, the Waste Isolation Pilot Plant (WIPP), New Mexico or DOE's planned repository at Yucca Mountain, Nevada.

In framing this study the committee also considered a more general set of factors that affect DOE's current and future options for dispositioning its excess nuclear materials: legal or programmatic agreements, the attractiveness of the material for theft, e.g., by terrorists, and cost. These factors are summarized in Table 2.1. Except for spent nuclear fuel (Chapter 4) and a portion of the heavier actinides (Chapter 7), there are no agreements to dispose the excess inventory as waste. Security measures to prevent theft are not new to DOE, which successfully protected its materials throughout the Cold War. The committee viewed security as a subset of the overall need for developing improved matrices for immobilizing nuclear materials. Incentives for research include the large costs for managing the inventory, safety, and the materials' scientific potential.

The committee concluded that the following options encompass the end points available and provide a framework for research for managing and dispositioning DOE's nuclear material:

- shorter-term storage for materials that have an identified use;

Table 2.1 Factors Affecting DOE’s Options for Disposing of Excess Nuclear Materials

Material	Agreements	Security Issues	Cost Perspective
Plutonium-239	In June 2000, the United States and Russia agreed that each country will reduce its inventory of excess weapons grade plutonium by about 34 metric tons (DOE, 2000c; NAS, 2000).	Pu-239 is a principal component in nuclear weapons. DOE's efforts to enhance security of its surplus Pu-239 have been guided by recommendations in a series of NAS reports (See Chapter 3).[1]	DOE estimated that the cost of managing its excess plutonium amounted to about 30 percent of its overall budget for managing excess nuclear materials in FY 2001.[2] Cost of converting excess plutonium into mixed oxide fuel is estimated to be about $3.8 billion over 20 years (Siskin, 2002).
Spent DOE nuclear fuel	The Nuclear Waste Policy Act of 1982 (and its amendments) require DOE's Office of Civilian Radioactive Waste Management to develop a geological repository for high-level radioactive waste and spent nuclear fuel.	Some spent DOE nuclear fuels contain quantities of Pu-239, U-235, or fission products that could make them attractive for theft. Applying the NAS spent fuel standard would require that DOE SNF be no more attractive for theft than commercial SNF.	DOE estimated that the cost of managing its excess SNF amounted to about 50 percent of its overall budget for managing excess nuclear materials in FY 2001. DOE projected a life cycle cost for managing and disposing of DOE SNF of about $5 billion (DOE, 1998a).
Cesium-137 and strontium-90 capsules	Plans to accelerate the Hanford site cleanup call for transferring the capsules to an on-site dry storage facility by 2007. Final disposition has not been determined.	Acts to disperse radioactive fission products might be contemplated by terrorists. The radiation from the Hanford capsules is so intense that theft is unlikely; dispersal from the storage facility itself is a concern.	DOE estimated that the cost of managing its "other" materials, including the capsules, amounted to under 5 percent of its overall budget for managing excess nuclear materials in FY 2001.
Depleted uranium	The McConnell Act (Public Law 105-204) required DOE to submit a plan for treating and recycling its DUF_6 (completed July 1999) and constructing conversion facilities. In 2002 DOE signed an 8-year contract for constructing and oper ating the facilities. Final disposition has not been determined	Depleted uranium is not a security concern.	For FY 2001 DOE estimated that the cost of managing all its excess uranium amounted to about 15 percent of its overall budget for managing excess nuclear materials (DOE, 2000c). Cost of the DUF_6 would be less than 15 percent. The recently awarded contract for converting the DUF_6 has an estimated value of $558 million (DOE 2002e).
Higher actinides	Facility closures require moving or disposing the stored materials. For example, closing the Savannah River Site F-canyon may require disposing stored Am-243 and Cm-244 as high-level waste.	Given their relatively small quantities these materials are not attractive for theft.	DOE estimated that the cost of managing its "other" materials, including the higher actinides, amounted to under 5 percent of its overall budget for managing excess nuclear materials in fiscal year 2000.

---

[1] The Committee on International Security and Arms Control (CISAC) of the National Academy of Sciences (NAS) found that dispositioned DOE plutonium would continue to pose a unique safeguards challenge unless it were rendered approximately as inaccessible for weapons use as the much larger and growing quantity of plutonium in spent fuel from commercial reactors. A series of three reports (NAS, 1994, 1995, 2000) proposed and developed a “spent fuel standard” for comparing the accessibility of dispositioned DOE plutonium with the accessibility of plutonium in spent fuel from commercial nuclear power reactors.

[2] DOE provided cost estimates for managing excess nuclear materials in its Strategic Approach to Integrating the Long-Term Management of Nuclear Materials (DOE, 2000c) only on a “percent of total” basis. Because excess nuclear materials are managed by several DOE offices, which budget this activity differently, more detailed cost data are not available

- longer- or indefinite-term storage for materials that do not have an identified use but cannot or should not be disposed;
- disposal in WIPP as transuranic waste;
- dispersion into high-level waste (HLW) tanks for processing and disposal along with the tank waste;
- disposal in a geological repository designed for SNF and HLW; and
- disposal as low-level waste.

Uncertainties in waste acceptance criteria to be developed at the disposal sites make many of the planned end points for nuclear materials appear to be outside of DOE's control. Therefore, there is need and opportunity for research to support both the primary disposition options and the development of alternatives.

Setting Priorities in EMSP Nuclear Materials Research

A salient characteristic of nuclear materials is their potential for unforeseen, beneficial future uses. There is a tension between the needs of today's milestone-driven decisions and the planning of a longer-term research program. Currently, for example, meeting programmatic milestones is being treated as a fundamental objective. It appears to the committee that research opportunities are being foreclosed by a perceived need to adhere to programmatic milestones when the programs themselves are changing (DOE, 2002a).

Several themes emerged in the course of committee discussions of information gathered during the site visits:

- Plans and priorities for dispositioning nuclear materials are being set based on a fairly narrow focus, predicated on program schedules for meeting short-term objectives, for example, facility closure and process selection. The significance of the program schedules or even the continuity of these programs is not necessarily commensurate with the consequences of the decisions being made, for example, loss of unique materials.
- Nuclear materials pose special problems and unique opportunities. For example, handling radioactive materials requires expensive facilities and trained personnel. Some materials present security risks, and all present potential toxicological and radiological risks. On the other hand, some irreplaceable materials may have unforeseen beneficial applications, including basic scientific research.

It is not easy for any DOE office to formulate clear objectives when multiple stakeholders' and technical experts' points of view must be addressed within a realistic schedule and budget. DOE has recognized this challenge and developed a standard for risk-based prioritization, which includes the following high-level objectives (DOE, 1998b):

- maximize accomplishment of mission,
- minimize adverse effects upon public health and worker safety,
- minimize adverse effects upon the environment,
- maximize compliance with regulations,
- minimize adverse/maximize desirable socioeconomic impacts,
- maximize safeguards and security integrity,
- maximize cost effectiveness, and
- maximize public trust and confidence.

Each of these objectives addresses a particular type of risk, for example, health, safety, environmental, economic. To help accelerate site cleanup, EM has announced that it will prioritize its work to reduce risks (DOE, 2002a).

Research to Reduce Uncertainty: The Value of Information

In instances where a program objective has been established, uncertainty in how well an alternative approach might meet the objective may diminish the apparent advantages of that alternative. The value of research for reducing the uncertainty associated with the alternative can be quantified by using the tools of decision analysis.

Given a carefully developed set of objectives and associated performance measures, a widely accepted way to decide among alternatives is to evaluate their performance with respect to a utility function that maps each alternative's performance into a number that can be used to rank the alternatives with respect to the decision maker's objectives and preferences. Once the expected utility of each possible alternative is determined, the alternative with the best utility is chosen for implementation.

One purpose of research is to reduce uncertainties in such decision models. Uncertainty can reduce the expected utility of the decision alternatives; therefore, research can, in principle, add value by reducing uncertainty. For example, uncertainty with respect to a safety issue could drive selection of a costly alternative in order to assure mitigation of a hazard that may not be real. Examples include selecting treatments for impure plutonium to preclude pressurization of storage containers (Chapter 3) and conditioning spent fuel to meet yet-to-be-developed repository acceptance criteria (Chapter 4). Elimination of this uncertainty would allow selection of an alternative with a better resource allocation. In general, each key technical uncertainty in a decision analysis represents one or more candidate research projects, perhaps an entire subfield of research. An upper bound on the value of a given research project is quantified as the difference in the expected utility of the preferred alternative, with and without the uncertainty in the corresponding element of the decision analysis. This is called the "value of information" (see, for example, Clemen, 1996).

Research to Inform Future Decisions

The DOE Office of Science's mission includes both research and construction and operation of facilities as top-level fundamentals. The four goals are to (1) maintain world leadership in scientific research relevant to energy (including environmental impact); (2) foster the dissemination of results; (3) provide world-class scientific user facilities; and (4) serve as a steward of human resources, essential scientific disciplines, institutions, and premier scientific facilities (Dehmer, 1998).

However, even such broad objectives are not sufficient for establishing a research agenda. Any selection process based on these objectives still tends to value a given research project only in the context of individual focused decisions. A more global view is needed to properly value research aimed at generating new knowledge. Moreover, a decision process using only these objectives tacitly assumes the permanence of technical and programmatic decisions made today, without making allowances for new information or changing circumstances. Such an approach devalues longer-term research. However, the committee found that most of EM's science needs are derived from current program plans and milestones. The EMSP has traditionally accorded high priority to research directed to focused, mission-oriented "gaps" identified by technology coordinating groups at the DOE sites.

The committee was guided in its deliberations by considering a different sort of objective, namely the objective of preparing to make more informed decisions in the future. This approach has been formalized in recent papers on risk assessment and decision making, especially in the context of climate change research. There are important analogies between climate change policy and DOE's management of nuclear materials. Both areas affect future generations as well as the present generation, and neither can be addressed optimally by a static decision model aimed at resolving all issues now, in the face of all of today's uncertainties. The main point is that there is a better approach than trying to settle policy now, for all time, in light of substantial uncertainties.

> Rather than attempt to model the long-term consequences of current decisions, analysts should use near-term proxy measures to describe the government's ability to deal effectively with future decisions when they are made. . . . [T]he framework outlined here . . . argues for an adaptive approach that focuses on selecting policies based on near-term consequences, *and the learning they will provide to place governments in better positions to address climate change decisions in the future* [emphasis added] (Keeney and McDaniels, 2001, p. 992).

In other words, for purposes of the near term, one supplements a preliminary set of fundamental objectives with the proxy objective "to position ourselves better to address these same fundamental objectives later on." This includes fostering intellectual capital, fostering institutional capital, and developing an improved basis for evaluating alternatives or for formulating better ones in the first place. This implies a program of research to position ourselves better in the future, and it declines to presume that current programmatic assumptions should foreclose certain kinds of research (see also NRC, 2003).

Framework for the Committee's Recommendations

In the spirit of the aforementioned considerations, the committee has identified the following proxy objectives that, together with the specific DOE programmatic objectives, drive the research recommendations.

- Develop and maintain intellectual capital. As in any field of science, research on nuclear materials requires special expertise. It is well known that expertise in many relevant subfields is being lost, for example, actinide chemistry. One important dimension of a research program is maintaining (even recovering) expertise in these subfields.
- Maintain critical facilities. Research with nuclear materials requires special facilities, e.g., for containment and often for remote operations. A substantial investment exists in certain kinds of facilities. This investment will be lost if these facilities are decommissioned. A snapshot of strictly near-term fundamental objectives might not provide a basis for maintaining critical facilities, but a longer-term view might lead to a different conclusion.
- Keep options open.
  1. Preserve unique materials. Certain materials were produced by repeated cycles of high-flux neutron irradiation followed by purification. Lacking an immediate use for these materials, they may simply be designated as waste. It is easy enough to see how irretrievable disposal of these materials rates favor-

ably in light of a set of objectives that prioritizes minimizing current hazard and "mortgage" costs; but once lost, these materials would be extremely difficult to re-create. Metaphorically speaking, these materials transmute from "waste problem" to "opportunity" depending on the set of objectives being considered.
    2. Do not foreclose fundamental research programs just because a current program plan seems to moot the expected results. At some sites, current milestones for disposition of certain materials seem to preclude research into the phenomenology of those materials. This approach seems to base research decisions having long-term consequences on programmatic conditions that are subject to change.

- Improve the knowledge base.
    1. More knowledge supports better evaluation of alternatives.
    2. Better alternatives might be forthcoming from an improved knowledge base.

EM, charged with cleaning up and closing sites across the complex, for very good reasons is focused on going out of business as soon as possible. Disposing of surplus nuclear materials as waste is the simplest expedient. However, DOE will continue to use and supply nuclear materials. Furthermore, given the fundamental constraints on energy production, there is a real potential for new developments in nuclear power. Maintaining the nuclear material resources in DOE's current inventory, as well as research investments to expand the knowledge base for their future beneficial application, were overarching considerations for the committee as it developed its research recommendations. Improvements in the knowledge base have downstream potential value that goes well beyond the current EM mission.

Conclusions

Research activities serve two fundamentally different kinds of objectives. EM's cleanup objectives require near-term solutions to specific current problems. A second kind of objective is to be better able to address future problems: to be able to formulate, analyze, and implement new alternatives that may be needed to address changing needs or make better use of new information. By more explicitly recognizing this latter objective, which is a proxy for today's unidentified longer-term needs, the EMSP can strengthen its research planning.

3

Plutonium-239

Making the plutonium isotope of mass 239 (Pu-239) was a principal objective of nuclear materials production in the United States from the 1940s through the late 1980s.[1] Approximately 100 metric tons of Pu-239 were obtained from the nuclear reactors and separations facilities at the Hanford, Washington and the Savannah River, South Carolina sites for use in nuclear weapons (see Chapter 2 and Appendix A). About half of this production has been declared as surplus.[2] The surplus inventory includes clean metal, mainly from disassembly of weapons; oxide; and plutonium combined with a variety of other materials in reactor fuels, targets, and miscellaneous forms (see Figure 3.1). From its fact finding, the committee believes that managing and disposing of Pu-239 is the Department of Energy's (DOE's) greatest nuclear materials challenge, and presents the greatest needs and opportunities for near- and longer-term research by the Environmental Management Science Program (EMSP) and other DOE programs.[3]

DOE's efforts to manage surplus Pu-239 have been guided by the recommendations of a 1994 National Academy of Sciences study of plutonium disposition. The study made four specific recommendations (NAS, 1994, p. 2):

1. minimize the time during which the plutonium is stored in forms readily usable for nuclear weapons;

---

[1] Plutonium is a metallic, chemically reactive element. It does not occur naturally on Earth, but it is produced in nuclear reactions. Tritium, another product of nuclear materials production, is not discussed in this report.

[2] According to publicly available information DOE manages approximately 99.5 metric tons (MT) of plutonium, mostly Pu-239, at eight sites. Of this inventory, 52.5 MT are excess to national security needs (DOE, 2000c).

[3] Other plutonium isotopes in the DOE inventory are addressed in Chapter 7 of this report.

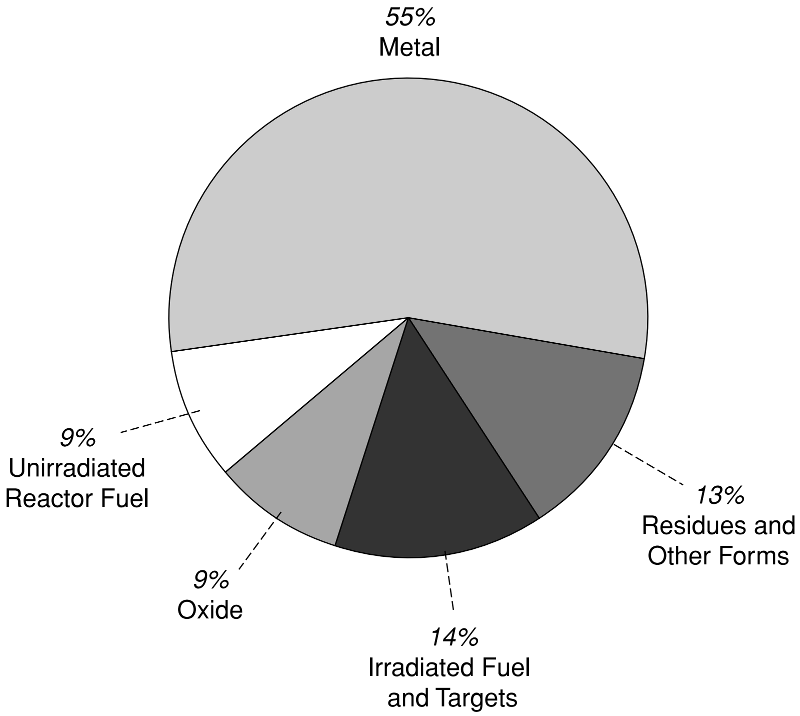


Figure 3.1 DOE has declared that about 50 metric tons of Pu-239 are in excess of national needs. About half is clean metal from disassembly of nuclear weapons and other sources. A variety of chemical and physical forms comprise the remainder. DOE plans to convert most of the excess Pu-239 to mixed oxide fuel for commercial power reactors. Source: DOE 2000c.

> 2. preserve material safeguards and security during the disposition process, seeking to maintain the same high standards of security and accounting applied to stored nuclear weapons;
> 3. [provide] a form from which the plutonium would be as difficult to recover for weapons use as the larger and growing quantity of plutonium in commercial spent fuel; and
> 4. meet high standards of protection of public and worker health and for the environment.

Currently efforts at most DOE sites are directed at gathering, processing, and storing excess plutonium according to the DOE 3013-2000 Standard “Stabilization, Packaging, and Storage of Plutonium-Bearing Materials” (DOE, 2000b). The standard provides for storage of stabilized, solid material in specially designed cans for up to 50 years (see Figure 3.2). Some sites, such as Rocky Flats, are being “de-inventoried” of their plutonium, which is being shipped to consolidate the inventory at a limited number of sites, such as Savannah River.

A key element in DOE’s strategy for eventual disposal of its surplus inventory is the conversion of as much of the excess Pu-239 as is technically and economically feasible into mixed oxide (MOX) fuel that can

Figure 3.2 DOE 3013 cans are intended to provide safe storage of excess plutonium for up to 50 years. Use of these containers will help DOE to "de-inventory" many of its facilities, close some sites such as Rocky Flats, and consolidate the plutonium at a few locations. The 3013 cans are typically 5 to 6 inches in diameter and 10 to 12 inches long. Source: DOE Los Alamos National Laboratory.

be used to generate electricity at commercial nuclear reactors.[4] The spent MOX fuel would be disposed with other DOE and commercial spent fuels, for example, in the planned Yucca Mountain repository. Most of the surplus plutonium is in the form of pits,[5] clean metal, and oxides for which conversion to MOX fuel should be straightforward, based on French experience (Johnson and Brabazon, 2002). A pit disas-

---

[4] The committee did not review the MOX fuel program.

[5] A "pit" is the core of a nuclear weapon.

sembly and conversion facility and a MOX fuel fabrication facility (MFFF) are currently being designed for construction at the Savannah River Site (SRS) (Siskin, 2002).

The inventory includes approximately 17 metric tons (MT) of plutonium in the forms of scraps and residues, including very impure materials with high halide content. Until canceling the project in early 2002, DOE intended to immobilize this impure Pu-239 in a ceramic material for co-disposal with vitrified high-level waste (DOE, 2000c). The immobilization facility was to have been built at SRS and used in conjunction with the planned pit disassembly facility, MFFF, and the currently operating Defense Waste Processing Facility, which is vitrifying high-level tank waste at the site. The immobilization facility was cancelled primarily for economic reasons (Siskin, 2002).

Coincident with canceling the immobilization facility, DOE modified the MFFF to include an "alternate feedstock" capability, which is intended to allow processing of most of the 17 MT. However, the technical basis for this plan remains uncertain. According to an assessment by the Defense Nuclear Facilities Safety Board (DNFSB), "While there is a reasonably good assurance of sound disposition paths for 36 MT of these [Pu-239] materials, the disposition paths for approximately 17 MT of metal, oxides, and fuel have considerable uncertainty. Of this latter group, about 6 MT of material has no disposition path" (DNFSB, 2002, p. 3-8).

Disposition Options and Challenges

DOE's disposition options for surplus Pu-239 include the following:

- storage according to the DOE 3013 Standard for up to 50 years;
- fabrication into MOX fuel;
- disposal as transuranic (TRU) waste in the Waste Isolation Pilot Plant (WIPP); and
- disposal along with high-level waste and spent fuels, for example, in the planned Yucca Mountain repository.

Extended Storage

At the end of the Cold War, the redirection of mission and scheduled closure of plutonium processing facilities in the DOE weapons complex left significant quantities of Pu-bearing materials in interim or ill-defined status. Normal processing operations would have moved these materials to final disposition or utilization. The DNFSB Recommendation 94-1 (DNFSB, 1994; DOE, 1994a) led DOE to establish a program to stabilize, secure, and process these materials to enable closure of plutonium facilities that no longer have a sustainable mission. This program continues to sponsor research and to develop and implement techniques to convert and stabilize plutonium-bearing materials for storage of up to 50 years. The program's goal is to have materials with greater than 30 weight percent plutonium in a stable, solid form that meets the DOE 3013 Standard.

The main challenge in implementing the 3013 Standard is reduction of moisture to within acceptable limits in the plutonium-bearing materials. The problem with moisture contained within radioactive solids is that in a confined, sealed package pressure can potentially build as a consequence of radiolytic decomposition of water and perhaps other molecules. In addition, gases produced in these sealed containers are expected to contain enough hydrogen gas to be of concern from safety and transportation standpoints. Plutonium-bearing materials that are less pure than metals and oxide frequently contain highly variable and poorly characterized constituents. These materials increase the challenge for processing to meet the 3013 Standard.

MOX Fuel Fabrication

For eventual disposition of the inventory, as much of the excess material as possible—including all materials currently in pit form—will be processed and fabricated into MOX fuel for once-through use in commercial nuclear reactors. The DOE has solicited specifications from European nuclear fuel manufacturers who routinely perform MOX recycling. Following use in commercial nuclear reactors, the spent fuel is expected to be packaged and shipped to Yucca Mountain (assuming it is licensed and constructed) for disposal.

The technical basis for MOX fuel has been fairly well established through work by DOE and its predecessor agencies (Freshley, 1973; Pedersen, 2002). The barriers to a substantial program are economic and institutional. First, the fuel fabrication facilities used for current light water reactors (LWRs) cannot accept MOX feed material because of radiation safety and safeguards requirements. The MOX facility ends up much more expensive in both capital and operating costs for equivalent throughput. Second, the fuel rods and assemblies will have much more stringent handling and shipping requirements than clean LWR fuel. Only when they reach the fuel canal of the reactor site will they be handled using techniques familiar to today's operating personnel, who are used to handling both clean and partially burned LWR assemblies in refueling operations. Third, the licensing basis for the fuel must account for the differences in reactor transient and control characteristics, accident behaviors and consequences, and safeguards requirements. The facility development, operational characteristics, and other important aspects related to developing a significant MOX program are currently being addressed by a number of research activities (Anderson, et al., 2002; Johnson and Brabazon, 2002; Naugle, 2002).

DOE has estimated that only about 2 of its 17 MT of impure plutonium-bearing materials cannot be sufficiently purified (for technical or economic reasons) to meet MOX feed specifications (Siskin, 2002). A representative distribution of some of these less pure plutonium materials, stored at Hanford (most of which were transferred from Rocky Flats, Colorado), is shown in Figure 3.3 (Venetz, 2002). The variety of materials, expressed as a function of plutonium content versus number of items, is from an inventory of about 900 items containing approximately 0.8 metric tons of plutonium with a total weight of 1.1 metric tons. Of note are the 79 items with plutonium contents less than 50 weight percent. In addition, there are also significant numbers of inventory items with plutonium contents ranging from 50 to 70 weight percent. Most of the analyzed samples have high chloride concentrations. Furthermore, many (if not all) of these materials have silicon concentrations ranging from 4 to 10 weight percent. The chemical properties of chloride and silicon, respectively, are detrimental to stor-

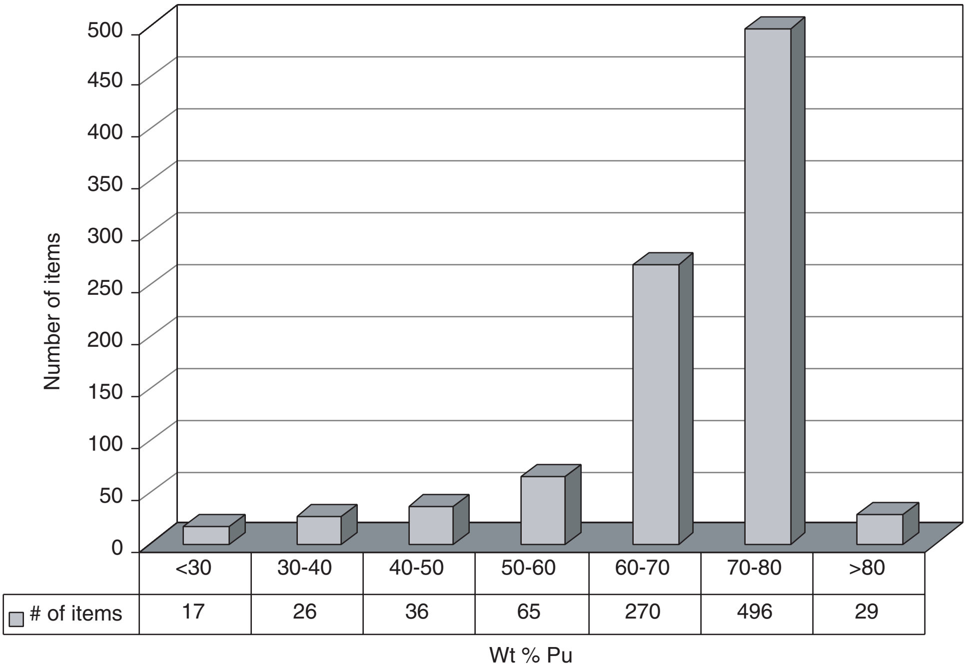


Figure 3.3 An inventory of impure plutonium oxides stored at Hanford shows that some materials may be too poor in plutonium for economical conversion to mixed oxide fuel but too rich for disposal as transuranic waste. Research may help improve both the technologies and the basis for decision making for dealing with such materials. Source: Venetz, 2002.

ing plutonium in the 3013 cans and processing it to make MOX fuel (WSRC, 2002).

Disposal in WIPP

Besides conversion to MOX fuel, DOE's other planned disposition route for excess plutonium is disposal as TRU waste at the WIPP in southeastern New Mexico. DOE currently has some 111,000 m^3 of TRU waste in retrievable storage awaiting shipment to the WIPP. Most of this volume consists of contaminated clothing, equipment, and process wastes that contain relatively small amounts of plutonium. Plutonium-bearing scraps and residues that meet the repository's waste acceptance criteria (WAC) can also be disposed. The WAC include nuclear safety limits of less than 200 "fissile gram equivalents" of Pu-239 per 55-gal drum of waste; no compressed gases, liquids, reactive or corrosive wastes; and no pyrophoric materials (DOE, 2002f).

As an example of increased WIPP utilization, DOE has recently amended a record of decision to allow an additional 0.97 MT of low assay oxides containing about 0.18 MT of plutonium to be sent to WIPP (DOE, 2002c, 2002d). These materials are similar to other plutonium residues that are already being sent to WIPP from Rocky Flats. Because of their impurities, DOE considers that these materials would be impractical to ship to SRS for fabrication into MOX fuel (see previous section). However, proposals to dispose of unanticipated large amounts of plutonium are likely to encounter political resistance (Domenici, 2002).

Co-disposal with Defense High-Level Waste

The DNFSB noted that there are uncertainties in DOE's current plans for its 17 MT of impure plutonium. The committee believes that as a practical matter some scraps and residues will fall in an ill-defined category of containing too many varied impurities for conversion to MOX and being too concentrated to meet the WIPP criteria. For these materials, DOE could revert to its earlier option of co-disposing some of this plutonium with defense high-level waste and spent fuels (NAS, 1994).[6] In the earlier option, incorporating the plutonium into a ceramic was proposed to meet waste acceptance criteria and provide resistance to theft (NAS, 2000).

The principal challenge in formulating crystalline ceramic hosts is radiation-induced transformation from a crystalline to an amorphous state (amorphization), mainly the result of self-irradiation damage asso-

---

[6] A review of research and development in support of this option is at http://www.llnl.gov/str/gray.html.

ciated with the alpha-particle emission (α-decay) of incorporated actinides, which can lead to large volume changes, increased surface area due to cracking, and enhanced corrosion rates. An acceptable host will have to accommodate at least a 10 percent loading of Pu to limit the overall waste volume, and remain stable for several hundred to several thousand years.

Radiolytic degradation of certain host ceramics, principally silicates and some phosphates, is an issue because cumulative ionization doses can reach 10^9 Gy in about 300 years for a 10 percent host loading of Pu-239, and $> 10^{11}$ Gy after 100,000 years. Over the past decade, DOE research efforts for plutonium disposition in crystalline hosts have focused on titanates, such as brannerite (UTi_2O_6), which are not as susceptible to radiolysis. Such structures eventually amorphize by accumulation of damage zones produced during α-decay by energetic recoil of the actinide nucleus (kinetic energy ~100 keV, range ~40 nm). Irradiation-induced amorphization competes with simultaneous recrystallization and typically occurs at a cumulative displacement dose of ~0.1 to 1 displacement per atom below a critical temperature that is often well above room temperature.

Research Needs and Opportunities

The EMSP should support research to help maximize the portion of DOE's excess plutonium inventory that can be used as MOX fuel and that will support the scientific basis for disposal of impure plutonium not suitable for MOX. Research should include fundamental chemistries for storing and purifying plutonium, modeling of MOX fuel performance to help ensure reactor safety, and devising high-integrity, theft-resistant forms for disposal.

There are opportunities for the EMSP to partner with other DOE offices (National Nuclear Security Administration, DOE Office of Environmental Management) to support plutonium storage and fabrication into MOX fuel. Ongoing projects in other offices could be extended to improve scientific understanding of long-term corrosion, gas generation, and moisture analysis relevant for storage of plutonium-bearing materials, process analytical chemistry and materials characterization for MOX fuel fabrication, and nondestructive assay technique improvements for use in high-radiation environments. Developing plutonium immobilization matrices that would resist illicit recovery of the plutonium or its dispersion is desirable.

For less pure materials, characterization and separation of undesirable impurities are needed to render more of the material suitable for MOX fuel. Research in support of MOX fuel fabrication programs may allow greater flexibility in incorporating a broader range of materials than current MOX fuel specifications allow. For material not destined for MOX fuel or WIPP[7] there are opportunities for research toward developing highly durable forms for co-disposal with high-level waste and spent fuel. Finally, there is potential for crosscutting research toward stabilization of both spent fuel and plutonium residues for storage, as well as for eventual disposal.

Stabilization for Storage and Transportation

The interim processing end point for materials with greater than 30 weight percent plutonium is a solid material that has been processed according to the DOE 3013-2000 Standard (DOE, 2000b). The transportation of stabilized plutonium-bearing solids follows from the requirements of the Code of Federal Regulations, Article 49, concerning transportation of containers bearing plutonium materials, that they not contain pyrophoric or potentially explosive contents (Bailey et al., 2000). These requirements are satisfied by processing and packaging the plutonium according to the 3013 Standard. The stabilization requirements and the container design represent a balance between achieving consistently low moisture content (to avoid container pressurization via radiolysis) and containing a "worst case" internal pressure. Surveillance programs have not, however, verified the high pressures predicted by the pressure equation currently used in the 3013 Standard (Berg et al., 2002; Duffey and Livingston, 2002).

There are opportunities for research on long-term corrosion and gas generation mechanisms inherent in these sealed, plutonium-bearing materials to ensure that they remain safe regardless of the duration of storage while reducing unnecessary conservatism in the standard. This research would involve fundamental study of atomic interactions on the solid surfaces of the stored material. Material forms of concern include those with relatively high salt residues, for example, plutonium mixed with residual magnesium and calcium compounds and other solids thought to be largely oxides and hydroxides, resulting from calcination

---

[7] Research needs and opportunities for plutonium-bearing materials designated as TRU waste and destined for WIPP have been described in previous reports (NRC, 2002).

in air environments at 950 °C. The hygroscopic property of these materials dictates that some moisture will be present following 3013 Standard canning in most plant operations.

Research into novel, noninvasive (in situ) pressure and hydrogen content-measuring technologies is of potential utility as a cost-efficient means to track and identify future problems in sealed materials in interim or long-term storage. In addition, there are crosscutting issues concerning interaction of water with uranium and spent nuclear fuel elements as well as with plutonium. For example, are the mechanistic details of gas generation common to many of these actinide surfaces? Are there molecular or material science issues that are unique to each element or material type?

Regardless of whether processed material is intended for conversion to MOX fuel or simply stored and secured for time intervals as long as 50 years, understanding the corrosion aspects of the storage system has high pragmatic value (Kolman, 2001). Corrosion may be increased due to elevated temperatures that are likely to occur during storage for two reasons: the multi-barrier 3013 storage canister tends to insulate the plutonium, and the poorer grades of plutonium are contaminated with isotopes (Pu-240, Pu-241, Am-241) that have shorter half-lives and thus produce more heat than pure Pu-239. Whether these materials can be stored at potentially elevated temperature remains an unresolved question and represents a gap in technical knowledge. Open questions regarding the combined effects of conventional radiation-assisted corrosion in these long-term storage environments need to be addressed through fundamental research programs.

There are other less fundamental, but nevertheless important, research opportunities to improve the plutonium calcination process. Furnaces and furnace components frequently exhibit spectacular and debilitating corrosion effects that seriously affect processing timelines, and secondarily, site closure schedules. These issues have been identified in disposition planning documents driven by specific site concerns (WSRC, 2002) but are common to many sites attempting safe processing and securing nuclear materials.

In addition, material protection, control, and accounting (MPC&A) are always of concern for plutonium-bearing materials to ensure that diversion or theft of weapons-grade materials does not occur. Although conventional nondestructive analysis techniques are frequently used to verify the fissile content of materials, there are opportunities for research into more efficient and rapid MPC&A technologies. As noted in Chapter 7, the nation's supply of Pu-244, used as a tracer in isotope dilution mass spectrometry for control and accountability determinations, is severely limited.

Conversion to MOX Fuel

DOE's decision to convert its 17 MT of impure plutonium to MOX presents a variety of research needs and opportunities. These "alternative feedstocks" will require processing and polishing operations to remove specific elemental and fission-product neutron poisons detrimental to their use in reactors and to remove constituents that are incompatible with long-term stability of fuel-rod cladding. Specific examples relating to conversion of weapons-grade plutonium include removal of gallium down to very low concentrations and removal of silicon to acceptable levels (SROO, 2002).

Techniques and methods for processing impure plutonium (scrap recovery) have been used successfully since the early 1950s (see Appendix A). The challenge is to extend these methods to economically recover increasingly impure portions of the 17 MT inventory. Along with research to help ensure cleanup of feedstock to established impurity limits, research that might lead to relaxing the limits for some impurities could have a substantial economic benefit.

Additional basic process engineering research is also warranted to convert plutonium materials into suitable MOX fuel in the most efficient manner minimizing cost, time, and facility usage. A specific question is whether multiple batch operations are a more efficient means of processing both routine and nonroutine feedstocks than continuous flow-through processing. Rapid low-cost analytical means of determining impurity levels of feed materials will be required to answer such a question. In addition, accurate determination of impurity levels in the highly radioactive environment of the plutonium matrix poses unique challenges for the analytical chemist. Current means of determining impurity levels of interest at sub-ppm levels require arduous dissolution procedures followed by atomic emission spectroscopy techniques. Improvements in this arena are needed to make rapid decisions about process efficiency.

Another key research area that would benefit MOX fuel processing includes nondestructive analysis (assay) methods (NDA). Improved methods can facilitate examination of nuclear fuel to assure that materials are not being diverted for clandestine purposes. Advantages of NDA techniques include remote operation and analysis, lower safety risk, and analytical determinations in difficult-to-assess or field environments. Nuclear safeguards and surety programs have historically provided the bulk of the funding to develop and implement these techniques, but there is opportunity for the EMSP to enhance the fundamental scientific basis for new techniques.

Alternatives for Co-Disposal with High-Activity Waste

The committee believes that there is sufficient uncertainty in current plans to convert all excess plutonium to MOX fuel or to dispose of it in the WIPP to warrant further research to support disposal in a geological repository designed for high-activity waste. Crosscutting research could also provide scientific support for disposing of spent reactor fuels, discussed in Chapter 4, and other actinide elements, discussed in Chapter 7.

Ceramic Matrices

Research into immobilizing plutonium from difficult-to-process materials classes is warranted from the standpoint of long-term stable disposition and may include development of novel material types and technologies. A comprehensive set of research recommendations for evaluating alternative crystalline wasteforms for plutonium disposition with respect to radiation stability emerged from an expert panel convened under the auspices of the DOE Council on Materials Science (Weber et al., 1998). The most promising actinide host phases cited include the structure types of pyrochlore, zirconolite, zircon, apatite, perovskite, titanite, monazite, zirconia, and sodium zirconium phosphate (NZP).

Stabilization of plutonium in crystalline solid forms is advantageous because the radionuclide is predictably coordinated in well-defined atomic sites in a durable solid that has been demonstrated—in many cases in geological analogues persisting over millions or billions of years—to resist release of the radionuclide in hydrologic corrosion conditions. Containment at well-delineated atomic sites in a crystal structure can supplement the geological barrier in providing assured isolation of very long-lived radioactive materials.

Crystalline ceramics may also be designed and synthesized to accommodate a specific radionuclide with optimal retention. Materials can be designed to obtain a desired radiation response, whether by building in certain defect structures to accommodate displaced atoms or providing ion sizes and charges and host structures that favor retention of crystallinity (and its predictable radionuclide coordination) when atoms undergo radiation disordering. It is the complexity and latitude of ceramics that makes these approaches possible and that provides opportunities for basic research. For example, it may be that the ideal radiation-stable plutonium host is an “amorphous” phase resembling the radiation-disordered phase that will eventually result and thus exhibit few further physical, chemical, or structural changes upon subsequent radiation disordering. Such an engineered phase might have predictable actinide coordination and short- and medium-range order little different from those of the crystal (Hobbs et al., 1999). Adding U-238 to the ceramic matrix may provide long-term criticality control by isotopic dilution as Pu-239 eventually decays to U-235.

Structural derivatives of the fluorite structure are potential crystalline hosts for plutonium because of the ability to readily incorporate actinides possessing fluoritic structures (UO_2, ThO_2, PuO_2). Furthermore, these structural types can readily incorporate ZrO_2 (of interest because of its use as an inert matrix for plutonium burnup in a reactor or accelerator-based neutron source). Additional structural types either under study or of potential interest include perovskites (e.g., $CaTiO_3$); zircon ($ZrSiO_4$); the silicotitanate sphene (or titanite, $CaTi(SiO_4)O$); the phosphate compounds monazite ($LnPO_4$); apatite ($Ca_{4-x}RE_{6+x}(SiO_4)_{6-y}(PO_4)yO_2$); NZP ($NaZr_2(PO_4)_3$); and the $A_2B_2O_7$ pyrochlores in which the A-site contains large cations (Na, Ca, rare earths, actinides); and the B-site smaller high-valence cations (Nb, Ta, Ti, Zr, Hf, Sn, Fe^{3+}, Pb). Zirconolite ($CaZrTi_2O_7$), a fluorite derivative closely related to pyrochlores, is the primary actinide host phase in the polyphase Synroc crystalline high-level waste form. Pyrochlores and related derivatives are especially attractive as long-term storage hosts because of the large range of elements (and actinides) they can accommodate, which is a particularly useful property in immobilizing plutonium from impure plutonium sources.

Recent studies (Sickafus et al., 2000; Wang et al., 1999) have shown that certain plutonium-accommodating titanate-based lanthanide pyrochlores can be synthesized and exhibit remarkable radiation resistance against amorphization by substituting Zr for Ti. Because pyrochlores are fluorite structure derivatives, disordering the cation sublattices yields an average defect fluorite structure; the fluorite structure class is the most resistant to amorphization because of the huge redundancy in structural constraints (Hobbs et al., 1996, 1999). The closer the initial pyrochlore structure is to the fluorite structure, the more stable the cation-disordered structure is toward subsequent amorphization; this observation is more pronounced for the pyrochlore lanthanide zirconates than for the lanthanide titanates, and particularly so for actinide substitutions.

Several other countries have ongoing research in crystalline ceramics. These provide opportunities for international collaborations. The Australian National Science and Technology Organization has a research program that is now several decades old, which continues to investigate titanate-based waste forms. The most recent efforts have been to develop a pyrochlore ($Gd_2Ti_2O_7$) for the immobilization of plutonium. In Russia, a number of institutes work on ceramic waste forms (many supported by DOE). The principal structure types investigated include zircon, zirconolite, pyrochlore, NZP, and garnet. British

Nuclear Fuels, Ltd., is funding a number of university centers to develop a variety of waste forms. Researchers at Cambridge are studying radiation effects in zircon (a possible host phase for plutonium), perovskites, and Zr-pyrochlores. China and Japan have generally modest programs on waste forms, mostly investigating titanate phases.

Under French law, research programs have until 2006 to provide the necessary data to support the development of a waste disposal strategy. At present, the crystalline waste forms under consideration have been narrowed to zirconolite, monazite, apatite, and a thorium diphosphate-phosphite. In addition, the French still maintain a very active fundamental research program on the properties of glass waste forms.

Alternative Processing

A recently developed process (INEEL, 2001) involves the treatment of plutonium-containing salts with an electro-dissolution step that transports and deposits uranium (and plutonium if desired) in a lithium chloride-potassium chloride eutectic salt. Periodically the salt is disposed as high-level waste due to accumulation of fission products and other impurities, including plutonium if it is not recovered. The salt is ground and hot-blended with a zeolite, and the mixture blended with glass particles and roasted. Process control variables include types and particle sizes of salt, zeolite, and glass, and temperatures of contacts and roasting conditions. It is important to know, however, if the identity of the LiCl-KCl residual components is retained in the final waste form. While the product appears to meet waste acceptance product specification criteria in leach and durability tests as produced, at short times or with low activity surrogates, it may not do so in the long term with the radiation field of Pu-239, particularly if the chlorides are retained as alkali halide compounds and do not dissolve into the zeolite or glass matrices. Alkali halides are perhaps the most radiation sensitive of all materials and undergo efficient radiolytic decomposition in an ionizing radiation field; substantial decomposition would be expected in repository conditions in several hundred years for a 10 weight percent plutonium loading. Even if the salt is dissolved into the glass or the zeolite, both will also undergo substantial structural changes from ionizing and displacive radiation fields of the plutonium α-decay, and their chemical durability will change. Long-term stability for any new waste form in a radiation field needs to be investigated.

Canister Integrity

Once a plutonium-bearing material has been classified as a waste and is processed into a stable form, the integrity of its containment system needs to assured. Radiation-induced corrosion and other potential release mechanisms need to be determined. Although preliminary needs and critical issues have been identified in a number of recent communications (Wang et al., 1999; Weber et al., 1998) additional experimental work will undoubtedly be required for alternative material forms. Dimensional changes in the solid waste form, due to potential amorphization of crystalline components or restructuring of any glass components, are one such issue for the integrity of the waste form itself, as well as integrity of the encapsulating container. Anisotropic dimensional changes are known to cause disaggregation of polycrystalline waste forms through grain boundary and transgranular fracture. This is less a problem for fine-grained material (submicrometer to micrometer grain sizes) but becomes a significant effect for larger grain sizes; Synroc, for example, is typically produced with 10–100 µm grain sizes. Measurements of internal stress developing with irradiation in accelerated tests with short-lived isotopes, for example using X-ray diffraction-based or Raman-based stress measurements used for assessing stresses developing in oxidation scales, need to be carried out. Processing methods for producing fine-grained polycrystalline assemblages of host phases incorporating appreciable plutonium or other actinides need also to be researched.

4

Spent DOE Nuclear Fuel

The Department of Energy (DOE) manages an assortment of spent nuclear fuel (SNF) types that vary in both materials of construction and in quantity. DOE spent fuel was generated in military and civilian reactor development, research, and fuel testing programs. The inventory also includes irradiated fuel and target[1] assemblies that were placed in storage when DOE stopped reprocessing nuclear fuel for production purposes in 1992. Altogether there are over 250 different fuel types that have different enrichments, fissile materials, cladding, and geometries (DOE, 2000c). The fuels range from the Hanford N-Reactor SNF (about 85 percent of the total, see Figure 4.1), to the "cats and dogs" stored at the Idaho National Engineering and Environmental Laboratory (INEEL), which approach 150 different types. Currently a major effort is underway to retrieve N-Reactor SNF—some of which is damaged or deteriorating—from storage in pools at Hanford's K-Basin, dry the fuel, put it in multi-canister overpacks, and place it in a newly constructed dry storage facility. Similar efforts to ensure safe interim[2] storage are underway at other sites (NRC, 1998). DOE has recognized that small quantity "orphan" SNF, which is not included in these programs, may become an obstacle to site closure (Chambers and Kiess, 2002).

The various DOE SNFs have been characterized broadly into categories based on fuel content, composition, and cladding material type,

---

[1] Most of DOE's nuclear materials were created in nuclear reactors through the capture of neutrons by various target isotopes, e.g., U-238 (see Appendix A). Using separate fuel (driver) and target assemblies increased production efficiency. DOE manages most irradiated targets as SNF. The committee does not distinguish between fuels and targets when referring to SNF.

[2] Interim storage is temporary storage that is begun before its eventual duration is known.

Figure 4.1 Fuel from the Hanford N-Reactor comprises 85 percent of DOE's spent nuclear fuel inventory. The fuel consists of metallic uranium assemblies (two concentric cylinders with zirconium-based cladding) that are 66 cm long and 5 cm in diameter. In fresh fuel, as shown here, the U-235 enrichment is 1.25 percent. Source: DOE Richland.

as shown in Table 4.1. While the heterogeneity of DOE spent fuel will make its long-term management expensive and complex, the total inventory of DOE spent fuel—approximately 2,500 metric tons of heavy metal (MTHM)[3]—amounts to only about 5 percent of the current inventory of commercial spent fuel from power reactors. Furthermore, the current inventory of about 44,000 MTHM of commercial SNF will approximately double to about 84,000 MTHM by 2020.[4] The quantities of DOE spent fuel are also small compared to the 340,000 cubic meters of high-level waste that resulted from reprocessing spent fuel and target materials, mainly at the Savannah River Site (SRS), South Carolina; the Hanford Site, Washington; and INEEL (DOE, 2001a).

Disposition Options and Challenges

DOE plans to dispose of its SNF along with commercial SNF and vitrified high-level waste in a repository at Yucca Mountain in Nevada. Because DOE has only recently begun to prepare a license application for Yucca Mountain, significant uncertainty exists in what the waste acceptance criteria will be for many of the categories of DOE spent fuel. Commitments for waste form characteristics made during the

---

[3] Quantities of nuclear fuel are reported in terms of the mass of heavy metals, principally uranium but in some cases including plutonium, used in their fabrication.

[4] See http://www.eia.doe.gov/cneaf/nuclear/page forecast/projection.html.

Table 4.1 U.S. Department of Energy Spent Nuclear Fuels

Group	Fuel Type	Typical Fuel	Total (MTHM)	Percent of total
1	Zircaloy clad naval	Surface ship/submarine assemblies	65	2.6
2	Plutonium/uranium alloy	Enrico Fermi Reactor (Fermi) Core 1 & 2	9	0.4
3	Plutonium/uranium carbide	Fast Flux Test Facility (FFTF) test fuel assembly	0.1	0
4	Plutonium/uranium oxide and plutonium oxide	Fast Flux Test Facility (FFTF) driver fuel assembly	12	0.5
5	Thorium/uraniurn carbide	Fort St. Vrain	26	1.1
6	Thorium/uraniurn oxide	Shippingport Light Water Breeder Reactor	50	2.0
7	Uranium metal	N-Reactor	2,100	85.0
8	Uranium oxide	Three Mile Island core debris	180	7.1
9	Aluminum-based fuel	Foreign research reactor pin cluster	21	0.8
10	Unknown	Miscellaneous	5	0.2
11	Uranium-zirconium hydride	Training, research, and isotope General Atomic (TRIGA)	2	0.1
TOTAL			2,470	99.8

Source: Duguid et al., 2002; BSC, 2001.

Yucca Mountain licensing phase will affect the accuracy required for characterizing the spent fuel isotopic and chemical composition prior to waste acceptance and the nature of any spent fuel conditioning that will be required. For security, it is expected that DOE SNF will have to meet the “spent fuel standard” proposed by the National Academy of Sciences, namely that through a combination of size, weight, intensity of radiation, and fissile material content the material would be no more attractive for theft than commercial SNF (NAS, 1995).

The management of DOE spent fuel will first involve its continued interim storage for another 10 years or more and transportation to facilitate the consolidation of the materials in a smaller number of DOE sites. Some residues from this spent fuel, such as corrosion products found in the K-Reactor Basin at the Hanford site, will be treated as transuranic waste and sent to the Waste Isolation Pilot Plant facility in New Mexico for permanent disposal (McKenney and Walton, 2001).

The handling and conditioning of spent fuel for disposal will also generate low-level radioactive waste that DOE must disposition.

While reprocessing DOE spent fuel could potentially create a source of fissile and target material for reuse in future nuclear energy production, the heterogeneity and complexity of most of the inventory would make it a significantly less attractive source material for commercial fuel fabrication than the much larger, homogeneous quantities of commercial spent fuel that will be in storage, should the commercial demand for reprocessing emerge. The highly enriched uranium (HEU) spent fuels in the DOE inventory do retain a significant investment in enrichment that could be recovered by reprocessing and down-blending to create low-enrichment power reactor fuel. However, this HEU has substantial concentrations of U-236, a neutron-absorbing isotope, and U-232, which has a gamma-ray-emitting daughter, thallium-208. The presence of these isotopes substantially complicates using DOE HEU in low-enriched uranium power reactor fuel.

Most types of DOE spent fuel have important characteristics that are different from commercial spent fuel, which comprises most of the waste intended to be disposed in Yucca Mountain. These are primarily differences in the chemical forms of the fuel, the cladding materials that encase it, and the isotopic composition of the fuel. The different characteristics affect the spent fuel's chemical stability and potential for gas generation, the decay heat generation and the potential for thermal damage under different storage and accident conditions, the potential for inadvertent nuclear criticality, the potential doses for workers, and the attractiveness of the material for theft. Assessments are further hampered when current instruments and records are inadequate to characterize the chemical and isotopic composition of the fuel sufficiently. The areas where the unique characteristics of the DOE spent fuels influence safety and security, and thus create opportunities for research, can be subdivided into three categories: (1) chemical and thermal stability, (2) nuclear criticality, and (3) material protection, control, and accounting.

Chemical and Thermal Stability

Commercial spent fuel consists of ceramic pellets contained in zirconium-alloy tubes, called fuel pins or rods, which are highly corrosion resistant due to the requirement for protracted operation in high-temperature water inside the intense radiation environment of a reactor core. Several classes of DOE spent fuel have lower chemical stability than commercial spent fuel. These include metallic fuels and fuels with aluminum cladding. The cladding of some of these spent fuel materials is already corroded, some severely (see Figure 4.2). Also included in this category is the damaged fuel that was recovered from the Three

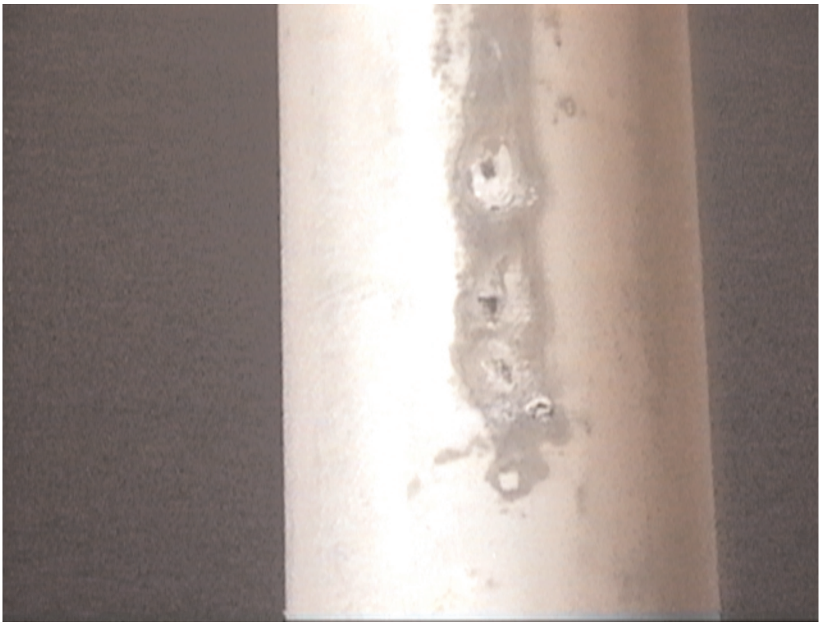


Figure 4.2 Most of DOE's spent nuclear fuel is stored underwater to provide cooling and radiation shielding. A variety of corrosion processes, such as the pitting shown here, are degrading many of the fuels. Source: DOE National Spent Fuel Program.

Mile Island Reactor. These materials have the potential to corrode more readily in both storage and repository environments and to generate radiolytic hydrogen during storage if not properly conditioned to remove water.

The fissioning or "burnup" of DOE SNF is lower but much more variable than that of commercial SNF, for which maximizing burnup for energy production is important economically. While the technical challenges of removing decay heat are thus lower, thermal safety analysis requires accurate characterization of the isotopic composition of the spent fuel. Conditioning to improve the fuel's stability also requires the capability to accurately characterize the chemical state of the fuel.

Nuclear Criticality

Several of the DOE spent fuel categories have significantly different criticality potentials during storage, handling, and disposal than commercial spent fuel, primarily due to differences in the isotopic composition of the fuel. Evaluations focus on worker safety because criticality can create dangerous radiation fields. Disposal regulations (10 CFR 63) require that criticality in a repository be considered and analyzed. For repository disposition, HEU spent fuels are the most important spent-fuel category that differs substantially from commercial spent fuel in criticality potential. The DOE HEU spent fuels include research-reactor spent fuel, naval spent fuel, and Fort St. Vrain gas-cooled reactor spent fuel.

Material Protection, Control, and Accounting

Several of the DOE spent-fuel categories contain weapons-usable materials in forms that are much more readily recovered than the plutonium and U-235 in commercial spent fuel. Commercial spent fuel has low uranium enrichment, relatively small plutonium concentrations (around 1 to 2 percent), and high radiation levels. These characteristics make commercial spent fuel an unattractive source of uranium for enrichment, and the recovery of plutonium is difficult and expensive. HEU spent fuel, and unirradiated or low-irradiation mixed oxide fuels containing plutonium, require more rigorous material protection, control, and accounting (MPC&A) to compensate for their lower intrinsic resistance to theft. Also, during any type of chemical stabilization or processing of spent fuels, accurate characterization of the isotopic and chemical composition of the fuel is required to permit accurate accounting for inputs and outputs from the process. These MPC&A issues have substantial similarities to the issues for the plutonium scraps and residues also considered in this report (see Chapter 3). A previous committee provided research recommendations to the EMSP for improving sensor technology and remote monitoring techniques (NRC, 2002).

Research Needs and Opportunities

The Environmental Management Science Program should support research to help ensure safe and secure storage and disposal of DOE SNF. Research should emphasize materials characterization and stabilization, including developing a better understanding of corrosion, radiolytic effects, and accumulated stresses. This research should be directed toward determining a limited number of basic parameters that can be used to evaluate the long-term stability of each of the types of DOE SNF in realistic storage or repository environments.

Material Characterization

The primary research challenge and opportunity in the area of characterization is nondestructive assay of plutonium and other radioactive isotopes in the high-radiation environment typical of most spent fuels. There are three areas in which characterization is necessary to ensure the suitability and stability of DOE SNF for long-term storage or disposal.

The first is to characterize the chemical and materials properties of the spent fuel to allow its stability for interim storage and disposition to be assessed. Important mechanisms that can degrade spent-fuel stability range from radiolytic gas generation to biocorrosion to matrix dissolution. Stresses can accumulate from the fuel's thermal history and from other effects such as swelling due to oxidation or radiolytic displacements and transmutations. Research is needed to identify parameters and methods of measuring them that better characterize the potential for these forms of degradation to occur.

The second is to characterize the isotopic composition of the fuel sufficiently to predict decay heat generation rates during storage and disposal, to assess the potential for criticality, and to provide an adequate description of the spent fuel to assess its ability to meet repository waste acceptance criteria. The isotopic composition is critical to the postclosure performance assessment of the repository, which is an essential part of the licensing case. For both commercial and defense spent fuel, the estimates of inventory depend on the fuel's in-reactor history, which may be poorly known. Consequently, conservative assumptions regarding burn-up and other factors may be required, but at the expense of an overly conservative design. If means were available to assay nondestructively the isotopic content of spent fuel, a potentially significant uncertainty could be reduced. The specific repository acceptance requirements for spent-fuel isotopic characterization may change as Yucca Mountain licensing progresses, and changes should be noted to ensure that appropriate characterization research is performed.

The third is to characterize the inventories of weapons-usable isotopes in the spent fuel to provide input for the materials accounting. Research is needed to develop improved methods to assay weapons-usable isotopes in spent fuel and could greatly facilitate materials accounting for fuel-conditioning methods that involve bulk processing of the fuel materials. Such methods could be tailored to the specific bulk-processing methodology. For example, the dry reprocessing methods being developed in South Korea to convert spent pressurized water reactor fuel into new fuel for heavy-water reactors provide a novel accounting method for plutonium. Neutrons from curium isotopes are measured at all processing stages. Because such dry reprocessing methods are incapable of separating plutonium and curium, the method allows accurate tracking of the plutonium inventory (Greenspan et al., 1998).

Stability in Storage

For spent fuels of relatively low chemical stability, such as DOE aluminum-clad fuels, some conditioning is likely to be needed to ensure that they remain stable enough to meet safe storage requirements until they can be emplaced in a disposal facility (NRC, 1998). A wide variety of potential degradation mechanisms exist: radiolytic gas generation, biocorrosion, pitting corrosion, interactions with other materials in storage containers, oxidation, matrix dissolution, and hydriding. There are opportunities for research to better understand these degradation methods and to identify inexpensive approaches to arrest them without resorting to more complex conditioning methods that would alter the entire fuel.

Stability for Disposal

Except for some corrosion products and ancillary low-level waste, the materials in DOE spent fuel are expected, ultimately, to be disposed in a deep geologic repository. What remains uncertain for many of the DOE waste forms is the degree of conditioning that will be required before the spent fuel can meet repository acceptance criteria.

In this context, if the waste acceptance criteria that ultimately emerge are quite broad, then the economically optimal disposition approach will be to dispose of DOE spent fuel directly to a repository, with minimum conditioning. Conversely, highly restrictive waste acceptance criteria might lead to reprocessing all of the DOE spent fuel to generate streams of high-level waste, plutonium, and uranium that are essentially identical to much larger quantities of commercial material the United States must manage in any case. The likely outcome is somewhere in between—some DOE spent fuels will require substantial conditioning, and others very little.

From the perspective of repository acceptance, minimal conditioning may prove to be problematic for HEU fuels, due to criticality issues, and for aluminum-clad fuels, due to chemical stability issues. Research is needed to identify and further develop conditioning methods that could facilitate repository acceptance. Further advances to the “melt and dilute” bulk-processing method, which reduces the criticality potential for HEU, would be an example. In this process, HEU spent fuel is melted down under carefully controlled conditions to drive off the most volatile and mobile fission products. The melt is then diluted with depleted uranium to prevent possible criticality and render the SNF unattractive as weapons material.

Research to further develop lower-temperature reprocessing options, where the spent fuel is dissolved in a molten salt or an aqueous solution, and separate streams of well-characterized materials are created, may help to address the specific issues of high enrichment and low cladding chemical stability that distinguish many DOE spent fuels from commercial spent fuel. There are opportunities for collaboration with the new DOE Advanced Fuel Cycle Initiative (AFCI) to identify research that would make the reprocessing approach viable for some DOE spent fuels that would otherwise have difficulty meeting repository waste acceptance criteria (DOE, 2003).

Developing a scientific basis for assessing the performance of DOE SNF in a repository environment and qualifying various DOE spent fuels for repository acceptance may be less expensive than reprocessing these spent fuels to create well characterized and understood waste streams. Such research will need to be integrated with ongoing repository licensing processes, to develop approaches to meet the waste acceptance criteria as these emerge, and to provide feedback to defining the waste acceptance criteria so these criteria do not inadvertently exclude materials for reasons that are not justified from the perspective of safety. Examples of appropriate research in this area could include the development of less soluble neutron poisons, and modeling of the interactions between spent nuclear fuel and waste glass if they are to be co-disposed in a single waste container.

5

Cesium-137 and Strontium-90 Capsules

In the early 1970s operators at the Hanford, Washington, site removed a large fraction of the Cs-137 and Sr-90 from the site's high-level tank waste in order to reduce the requirements for cooling the tanks. The cesium and strontium were concentrated and sealed in stainless steel (SS) capsules (see Figure 5.1) for potential uses, for example, thermoelectric generators and sterilizers. The expected applications for the Hanford capsules did not materialize and ceased entirely in 1988 after one capsule being used in the commercial sector was found to be leaking (USNRC, 1989). The almost 2,000 capsules contain a total of 67 million curies of radioactivity, which amount to about 37 percent of the total radioactivity at the Hanford site (ROO, 2002). They have been described as the nation's most lethal single source of radiation other than inside an operating reactor (Long, 2002). According to a recent letter of intent among the state of Washington Department of Ecology, the U.S. Environmental Protection Agency, and the Department of Energy (DOE), dispositioning the capsules is among eight specifically identified priorities for accelerating Hanford Site cleanup (Tri Party, 2002). Currently the capsules are stored at the Waste Encapsulation and Storage Facility (WESF) under 13 feet of water to cool the capsules and provide radiation shielding. DOE plans to move the capsules to passive air storage; however, their final disposition has not been determined (DOE, 2002g).

In the capsules, cesium is in the form of cesium chloride (CsCl) and strontium as strontium fluoride (SrF_2). The chemical composition has been described as being relatively uniform (NRC, 1997b). Each cesium capsule contains on average approximately 35,000 Ci of Cs-137 plus an unspecified amount of Cs-135 estimated to be 0.7 Ci and produces approximately 190 W of heat. Each strontium capsule contains approximately 33,000 Ci of Sr-90 and produces approximately 260 W of heat (see Table 5.1).

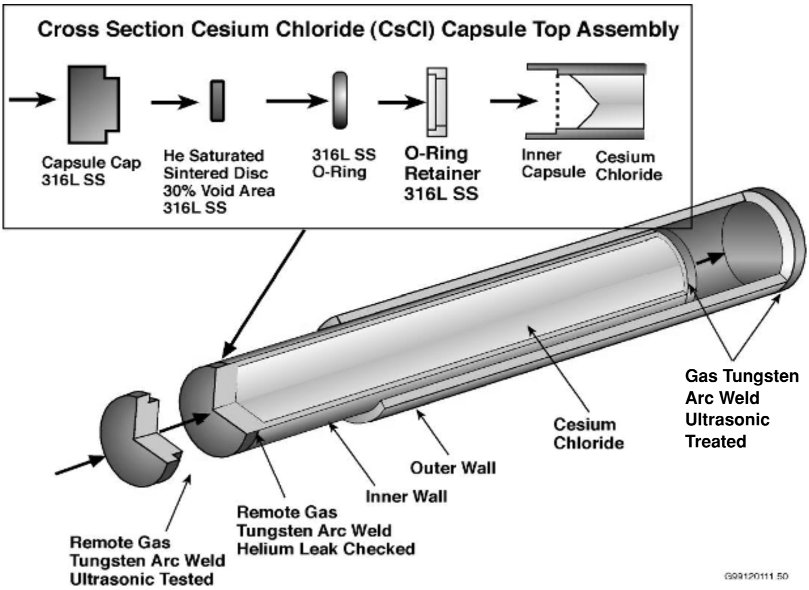


Figure 5.1 A typical Cs-137 capsule stored at Hanford may contain up to 40,000 curies of radioactivity. The capsule is made of 316L stainless steel with outside dimensions 20.8 inches long and 2.6 inches in diameter. Source: DOE Richland.

Table 5.1 Summary of 1995 Cesium and Strontium Capsule Inventory at the Hanford WESF Facility

Current inventory[a]	1,312 Cesium capsules 23 Cesium capsules in overpacks 601 Strontium capsules
Capsule dimensions[a]	Diameter (outside): 2.6 inches Length (outside): 20.8 inches
Capsule materials of construction[b]	Cesium inner and outer capsule: 316L SS Strontium inner capsule: Hastelloy C-276 Strontium outer capsule: 316L SS
Total volume[c]	Cesium capsules, 3.5 m^3 Strontium capsules, 1.5 m^3
Total mass[d]	15 Metric tons of capsules and contents
Radioactivity (as of 8/1/01)[a]	Cesium, 47 MCi Strontium, 20 MCi Total including daughter isotopes, 131 Mci
Heat generation (as of 8/1/01)[a]	355 kilowatts (1.2 million BTU/hour)

Sources: [a]ROO, 2002; [b]DNFSB, 1996; [c]NRC, 1997b; [d]DOE, 1996b.

Disposition Options and Challenges

The main challenges for managing and dispositioning the capsules are due to the intense radiation and the relatively large amount of heat that they produce. Dose rates range from 8,600 to 18,000 rems/hour for the Cs-137 capsules and from 20 to 420 rems/hour for the Sr-90 capsules.[1] Compared to other nuclear materials in DOE's inventory, Cs-137 and Sr-90 have relatively short half-lives, 30 years and 29 years, respectively. However, the present radioactivity is so great that it will take over 800 years for the Sr-90 to decay to the level currently allowed for disposal as low-level waste, 0.04 Ci/m^3. Cs-135, which is present along with the Cs-137, has a half-life of about 2 million years, and it will become the dominant source of radioactivity in the cesium capsules in about 600 years. Cesium and strontium have limited mobility in the environment due to adsorption on clays and other aluminosilicates. However, unlike most other elements, cesium can biomagnify (concentrate) as it moves to higher trophic levels (Whicker and Shultz, 1982).

Capsule integrity is essential for interim storage. Nevertheless, there are gaps in the technical basis for predicting incipient failures and understanding failure mechanisms. Most capsules are considered in good condition, but 23 have had to be overpacked, i.e., sealed in a larger stainless steel container (ROO, 2002). The need for overpacking is typically determined by simply shaking the capsule and listening for the inner capsule to "clunk." If the inner capsule moves freely, the capsule is deemed to be in good condition. However, if the inner capsule is swollen, it will not move freely and will be a candidate for overpacking. Various mechanisms for capsule failure have been proposed including poor welds and phase changes in the material as a function of temperature (DNFSB, 1996). The report states that the capsules "may have experienced chloride-induced stress corrosion cracking near the outer capsule welds due to lack of water chemistry requirements and control."

DOE's current plans (DOE, 2002g) include continuing to provide safe and compliant storage as well as surveillance and maintenance

---

[1] These are calculated dose rates at 3 feet from a bare capsule based on the contents of individual capsules (Source: Sen Moy, DOE Richland). The radiation is sufficiently intense that the capsules are self-protecting against theft, although the storage facility could be a target for the purpose of scattering radioactive material. The capsules themselves would likely meet the NAS "spent fuel standard" for resisting diversion (see Chapter 3 and NAS, 2000). It is recognized that Cs-137 and Sr-90 from smaller medical or industrial sources could be stolen for the purpose of dispersing radioactive contamination (IAEA, 2003).

activities for encapsulated cesium and strontium material, and continuing to maintain the WESF and structures associated with capsule storage. Several options are under consideration for disposition of the capsules. These options include

- continued storage in the pools at the WESF facility,
- passive storage in air at a new facility,
- overpacking and disposal of the capsules at a geologic repository, and
- vitrification into a glass or calcination into an oxide followed by disposal at a geologic repository.

Storage

Intermediate or long-term storage on site has the advantages of allowing monitoring and surveillance, providing physical protection, saving the material as a potential resource, and maintaining the material in disposal-ready condition while avoiding interstate transportation issues. Given their approximately 30-year half-lives, the isotopes could decay significantly during storage thus reducing their hazard and difficulty of eventual disposal. Issues for long-term storage include the commitment to maintain the storage facility and the capsule failure risk due to a lack of understanding of the processes occurring in the capsules.

For dry storage, the capsules would be moved using robotics and stored in air in a special facility designed to convectively exhaust the heat generated. In plans to accelerate the Hanford Site cleanup, the move from wet to passive dry storage is presented as a near-term goal (DOE, 2002g). However, the *Tri-City Herald* reported that it is unclear the state will agree to the dry storage concept (Herald, 2002). In any event, the dry-storage concept must be demonstrated viable and safe by addressing the aforementioned storage issues.

Disposal

Challenges for permanently disposing of the cesium and strontium involve meeting waste acceptance criteria at an appropriate facility. Under current regulations these materials cannot be disposed in the Waste Isolation Pilot Plant because they are not transuranic isotopes. They exceed U.S. Nuclear Regulatory Commission limits for shallow land disposal, which leaves disposal into the planned geological repository at Yucca Mountain as the only foreseeable option. At a vitrification workshop convened by the National Academy of Sciences in 1997, vitrification was reported to be “likely if overpack is unacceptable” for geological disposal (NRC, 1997b). Conversion to ceramics also appears attractive for stabilizing these chemically pure materials in a form that would resist dispersion.

Research Needs and Opportunities

The Environmental Management Science Program should support research that will help ensure continued safe storage and potential use or eventual disposal of the Hanford Cs-137 and Sr-90 capsules. Research should lead to understanding potential failure mechanisms of the present capsules, ways to convert the isotopes to stable glass or ceramic forms, and understanding long-term hazards of disposition options.

Stability of the Encapsulated Isotopes

There are research opportunities to understand the chemical and physical behavior of CsCl and SrF_2 under intense radiation and localized heating. There is little information for understanding the alterations that accompany the change of valence states during radioactive decay. Cs-137 (monovalent) decays into Ba-137 (divalent), and Sr-90 (divalent) decays into Zr-90 (normally tetravalent) via a short-lived Y-90 intermediate (NRC, 1996). Each of the decay products has very different physical and chemical properties (e.g., melting and phase transition points, ionic radius). Additionally, ionization due to the intense radiation fields is likely to induce other changes.

Topics for research include:

- the physical and chemical changes that will occur in the CsCl and SrF_2 in a confined environment (e.g., in sealed capsules) and intense radiation field,
- the physical and chemical changes caused by short-lived isotopes decaying to other elements with different valencies,
- the moisture and other impurity concentration thresholds to keep gas generation to acceptable levels, and
- the phase changes for these materials as a function of the impurity levels typical in these capsules.

Capsule Integrity

The DOE Office of Science and Technology's Multiyear Program Plan for FY02-05 (DOE, 2001b) includes two broad research categories for the capsules: (1) capsule integrity assessment methods for WESF at Hanford, and (2) a Cs/Sr capsule leak detection system for WESF.

Beyond these empirical tests there are opportunities for more fundamental research to help assure capsule integrity.

Factors that can affect capsule integrity include defects caused by intense radiation and heat-induced stresses, volume changes due to structural changes as the radioisotopes transmute, volume changes due to phase changes caused by chemical composition and/or temperature changes, and corrosion. Thermal effects will probably be more significant for dry storage since heat convection will be different than in the current water storage. This could induce repeated freeze-thaw cycling in the capsules if the temperatures were near a phase-change temperature. CsCl undergoes a phase change at 469 °C and melts at 645 °C (DNFSB, 1996). At first glance, the phase-change temperatures may seem to be acceptably high, but the presence of only 3 percent $FeCl_3$ causes CsCl to melt at 270 °C. Also, chloride and fluoride attack of the capsule material increases at higher temperatures.

Opportunities for research on capsule integrity include:

- developing more quantitative capsule surveillance methods than the "clunk" test to determine capsule material and weld integrity or bulging;
- understanding the fundamental reason for the capsule failures to date, specifically, the mechanical and metallurgical aspects, and applying the knowledge gained to assess the remaining capsules;
- understanding capsule content phase changes and/or chemical reactions to enable predictions about their behavior if the capsules are moved from storage in pools to passive air storage;
- understanding the gas-generation potential of the capsule contents over time, especially as the elemental composition changes dramatically in a matter of decades; and
- determining whether the capsule contents have a unique enough radioisotope signature to be able to identify them in the event of theft.

Matrices for Immobilizing the Isotopes

Unless disposing the capsules in simple overpacks can meet waste acceptance criteria, (e.g., at Yucca Mountain if licensed and constructed), it seems likely that immobilizing these isotopes in a durable matrix will be necessary—either for reuse or disposal. Options include vitrification in glass or incorporation into a crystalline ceramic. Because the cesium and strontium are essentially pure there is opportunity to stabilize these elements in tailored ceramics that are extremely durable. It also seems advantageous to maintain these isotopes in relatively small volumes rather than, for example, to attempt to blend them into the million-gallon volumes of high-level waste (HLW) in the Hanford tanks for vitrification. However, a long history of basic and applied research supports the vitrification option (Clark et al., 1986; DOE, 1994b). At this point it appears that there is not yet sufficient information to determine whether specialized waste forms or dilution into high-level waste glass is the better disposition path, and there is a clear need for research to help determine the better method.

Although the Hanford capsules appear to be theft resistant, immobilization research would also be applicable for stabilizing cesium and strontium from unused industrial and medical sources worldwide (IAEA, 2003). The chemical and physical form of the radioactive material would have a large effect on its dispersibility. If the cesium were in the form of an appropriate ceramic or other material that was insoluble and that broke up under high strain rates into nonrespirable large particulates (break along grain boundary with grain sizes above the respirable size), its potential for use by terrorists would be greatly reduced.

Research questions for immobilizing the isotopes in either glass or crystalline ceramics include the following:

- Does the cesium or strontium mix uniformly into the matrix or is there a tendency to segregate?
- What are the relevant phase transitions, especially with respect to volatilization at the elevated temperatures required to make either glass or ceramics? For example, impurities appear to reduce the phase-transition temperatures of CsCl.
- Can these salts be vitrified starting with the solid rather than from an aqueous solution?

Hanford's current plans are to vitrify its high-level tank waste. If the contents of the capsules are uniformly blended with the tank waste, the heat from radioactivity in the vitrified waste product will be increased about 50 percent. (As noted previously the radioactivity of the capsules is about 37 percent of the total radioactivity at the site.) If the blending is not uniform, then some glass with even higher thermal loads will result. High temperatures are detrimental to stability of glass (NRC, 1997b, 2001a), so additional research is needed to ensure long-term stability, especially since the purpose of the glass for immobilizing tank waste is to help sequester long-lived actinides for thousands of years.

Two principal issues for accommodation of cations in crystalline phases are transmutation and radiation damage. With respect to the first, the coordination of oxygen around cations, which stabilizes a given oxide crystal structure, depends on both ion charge and ion size, both of which can change upon transmutation. Monovalent Cs-137 transmutes to divalent Ba-137m and then to Ba-137, accompanied by a

20 percent decrease in ion radius, while divalent Sr-90 transmutes first to trivalent Y-90 and then to tetravalent Zr-90, with a 29 percent overall decrease in final ion radius. There has been little research on predicting the potential destabilizing effects of such cation valency and size changes in insulating crystals, particularly at high isotope loading.

Research is needed to develop novel ways to assess the structural stability of a given crystal type and ways to immobilize cesium and strontium. For example, in the structural tunnels or cages of crystalline zeolitic aluminosilicates or silicotitanates, where changes in ion charge and size may have less structural influence, the Si/Al or Si/Ti ratios could be adjusted to anticipate charge alterations over time to maintain charge neutrality.

The second issue is radiation damage. Radiation fields for both isotopes are extraordinarily intense. It is well known that the CsCl and SrF_2 solids currently in storage are acutely susceptible to radiolytic atom displacement (radiolysis). These halides undergo partial radiolytic decomposition to colloidal metal particles and evolvable halogen gas in the temperature range 100-200 °C after accumulated ionization doses in the dose region 10^8-10^{10} Gy. Therefore, research opportunities exist to study the stability of the waste type chosen given these large radiation fields.

Opportunities include research to:

- assess whether or not the current capsule form is viable for either long-term storage or direct disposal in a geologic repository,
- assess the merits of ceramic versus vitrified waste forms, including processes to produce them,
- develop methods for uniformly dispersing radionuclides in glass or ceramic and methods for verification,
- understand how to ensure glass or ceramic stability for time periods of hundreds of years under the intense radiation fields without corroding or leaching, and
- understand the physical and chemical changes that will occur for these very rapidly decaying compounds in a confined environment (sealed capsules, vitreous or crystalline matrices) under intense radiation fields and with substantial transmutations.

6

Depleted Uranium

Uranium is the parent of the Department of Energy's (DOE's) nuclear materials. The slightly radioactive element occurs naturally in the Earth's crust at an average concentration of about 2 parts per million. Minerals such as uraninite, pitchblende, and coffinite are principal sources of uranium. The uranium isotope with atomic mass 238 (U-238) comprises over 99 percent of natural uranium, 0.7 percent is the fissile isotope U-235, and there are traces of other isotopes. Operations to concentrate the U-235 isotope (enrichment) resulted in the accumulation of a large quantity of residual uranium depleted in U-235, referred to as depleted uranium (DU). The principal challenges facing DOE in dispositioning its DU arise from the large amount of material in the inventory and its chemical hazards.

Most DU is in the chemical form of uranium hexafluoride (DUF_6) amounting to 450,000, 198,000, and 56,000 metric tons, respectively, stored at DOE sites near Paducah, Kentucky; Portsmouth, Ohio; and Oak Ridge, Tennessee. There is a smaller but growing quantity under the responsibility of the U.S. Enrichment Corporation, which is still producing enriched uranium at Paducah for power reactors. The DUF_6 is stored in cylinders stacked in open-air storage yards (see Figure 6.1). Each contains about 14 tons of DUF_6. The Oak Ridge site has the oldest of these cylinders, some dating back to the Manhattan Project (Philpot, 2002). The most immediate risk posed by the DUF_6 is its potential to react with moisture to form hydrogen fluoride (HF), which is a highly corrosive and toxic gas, and uranyl fluoride (UO_2F_2), which is soluble and toxic.

At normal atmospheric pressure and temperature up to about 125 °F DUF_6 is a solid material (see Figure 6.2). Above 125 °F, a temperature that can be exceeded in the storage yard under direct sunlight, the material converts directly from a solid to a gas (sublimes). The density

Figure 6.1 Cylinders containing about 14 tons of depleted-uranium hexafluoride are stored outdoors. These cylinders are made of carbon steel with wall thickness of 1/4 or 5/16 inch. The stacking arrangement often prevents worker access for inspection or maintenance. Source: http://web.ead.anl.gov/uranium/guide/.

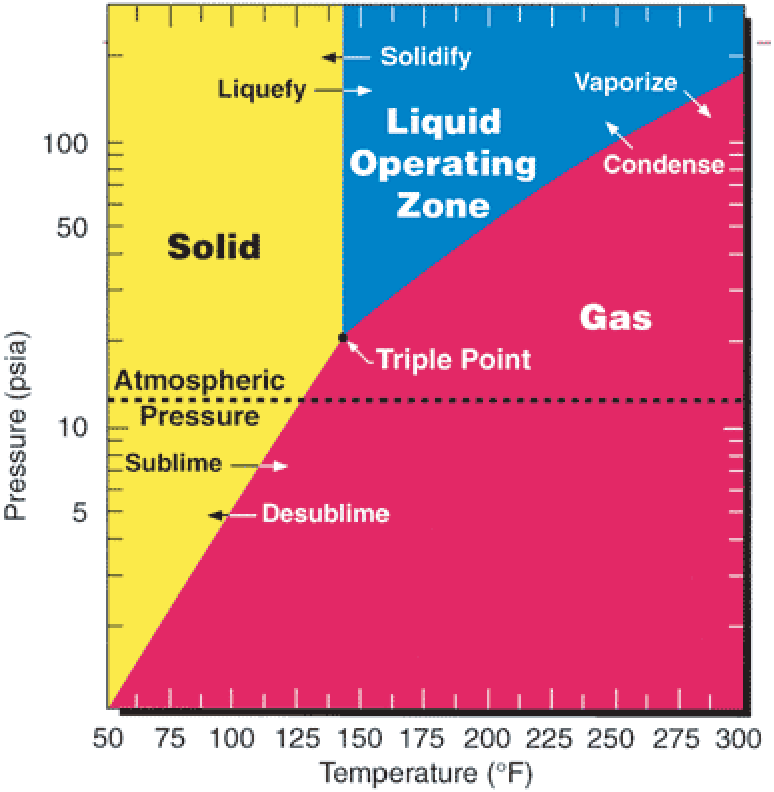


Figure 6.2 Phase diagram for uranium hexafluoride (UF_6) shows that the material can sublime—convert directly from a solid to a gas—under normal atmospheric pressure. Source: http://web.ead.anl.gov/uranium/guide/.

differences between the phases lead to a requirement that the cylinder contain enough ullage (free) volume that it can accommodate the phase transitions under extremes of temperature in the storage yard. A large release of DUF_6 would present a significant chemical hazard to workers and possibly to off-site populations.

In practice, examination has revealed cases where leaks have occurred through loss of integrity of the cylinders. In such cases, the leakage has not been sudden or large, as the leakage points tended to be self-healing due to buildup of uranyl (UO_2^{2+}) compounds from moisture reactions (see Figure 6.3). Carbon steel plates have simply been welded over the area as a patch. DOE considers such storage to be safe, but temporary.

DOE has recently taken a first step toward dispositioning its DUF_6 by awarding an 8-year contract to Uranium Disposition Services to build and operate facilities at Paducah and Portsmouth to convert it to the stable oxide U_3O_8 (DOE, 2002e). The Portsmouth plant will also convert the Oak Ridge DUF_6. The contractor will store the oxide at the two conversion facilities. A decision on final disposition of the DU has not been made. This recent action by DOE is consistent with studies that have concluded that conversion to the oxide would be the preferred course of action for the U.S. inventory (Haire, 2002). Abroad, the French company Cogema is proceeding with conversion to oxide at its enrichment plant. The Urenco partners (British, German, and Dutch national corporations) are continuing to store UF_6 cylinders at

Figure 6.3 Corrosion has breached the walls of some cylinders. Breaches have tended to be self-sealing. They are repaired by welding a patch over the opening. Source: http://web.ead.anl.gov/uranium/guide/.

Capenhurst, UK; Almelo, Netherlands; and Gronau, Germany. The worldwide inventory of DU is estimated to be about 1.2 million MT (NEA, 2001).

Disposition Options and Challenges

Options for future disposition of the DU, once converted to oxide, are continued storage, reuse, or disposal as waste. There are significant gaps in understanding health effects of uranium and its compounds that need to be resolved before DOE can fully evaluate these options. Beneficial ways to reuse large amounts of uranium have not been identified. Because of uranium's unique chemical and physical properties, the committee believes that this lack of reuse options reflects gaps in current knowledge rather than being an a priori reason for disposing of the material as a waste. There are significant challenges for deciding how the uranium might be disposed if it were declared to be waste.

Potential Health Issues

Surprisingly there are still substantive gaps in knowledge of the non-radiological health impacts of exposure to uranium and its compounds. For example, uranium can cause kidney damage in humans, but information on thresholds and the degree or reversibility of such damage is not available. According to the World Health Organization (WHO), studies in humans performed some 50 years ago cannot be used in quantitative risk estimation because the information on exposure, both qualitatively and quantitatively, is inadequate (WHO, 2001). Recent concerns over health effects of DU used in munitions have led to a resurgence of research in this area (Abu-Qare and Abou-Donia, 2002; McDiarmid et al., 2001; Miller et al., 2002).

Quantitative risk estimates for insoluble uranium have been obtained, albeit with uncertainty. Chemical dose-response and dose-effect relationships have not been investigated to any large degree. The extrapolation of data from animal studies to human response has not been investigated fully and a large degree of conservatism may exist. In fact, there appear to be differences in the sensitivity of different biological species to uranium toxicity, but no general picture has emerged. For humans, only very limited information is available on the inter-individual variation in uranium toxicity.

Reuse

The primary challenges to developing new uses for DU are lack of a scientific understanding of its potential health effects and public concern about its health effects. For example, uranium alloys and compounds are important in some materials applications because of their very high density ($\rho \sim 20$ g/cm^3). Depleted uranium has been used for shielding radioactivity and in armor-piercing projectiles for this reason.[1] It has also been proposed for counterweights or ballast in aircraft components (e.g., ailerons). However, concerns over the uranium's low-level radioactivity and toxicity have discouraged more widespread utilization.

Finding uses for relatively small quantities of DU will not solve DOE's disposal problem, nor would a use that would simply result in a different disposal problem. A major usage such as high-level waste repository overpacks might provide both a reuse and a disposal route. One proposed large-scale use for depleted uranium is as a component of multipurpose casks for commercial SNF storage, shipment, and disposal—one cask would serve all functions. DU can provide shielding and possibly enhance repository performance. For example, commercial SNF is made of UO_2. If it is surrounded by DUO_2, the DUO_2 can act as a sacrificial material to delay the degradation of the SNF under almost all conditions. The DUO_2 also reduces the potential for criticality in the very long term, after engineered barriers have failed, by isotopic dilution of U-235.

Disposal

The current plans for conversion to oxide will put the DU in a form that will be more stable than the DUF_6 for further storage. If disposal is necessary, it is not likely to be simple. The alpha activity of DU is 200 to 300 nanocuries per gram. Geological disposal is required for transuranic waste with alpha activity above 100 nanocuries per gram. If uranium were a transuranic element, it would require disposal in the Waste Isolation Pilot Plant (WIPP) based on its radioactivity. The chemical toxicity of this very large amount of material would certainly become a problem as well.

One option suggested by the U.S. Nuclear Regulatory Commission (USNRC) is disposal in a mined cavity or former uranium mine (Leeds, 2000). Challenges for this option would include understanding the fundamental differences between uranium ore (see Sidebar 6.1) and the bulk uranium oxide powder.

---

[1] Munitions fired in Kosovo in 1999 totaled about 9 tons of DU. This has raised new environmental and health concerns (Stone, 2002).

SIDEBAR 6.1 THE ENVIRONMENTAL BEHAVIOR OF URANIUM

Uranium's average natural concentration is approximately 2 ppm in the Earth's upper crust, where it exists in both the uranium(IV) and uranium(VI) oxidation states. Uraninite, UO_2, is the most widespread uranium mineral and the only common one with uranium in the tetravalent form. Pitchblende, UO_2, a major ore of uranium having the same idealized formula as uraninite, always contains some uranium(VI). Thus, a more realistic formula is $(U(IV)_{1-x}U(VI)_x)O2_{+x}$. Natural pitchblendes will have a stoichiometric range of uranium:oxygen from UO_2 to $UO_{2.6}$. Uraninite and pitchblendes vary chemically as a function of whether they are pegmatitic or magmatic uraninite or hydrothermal pitchblende. The first usually exhibits large amounts of rare earths and thorium, while the second does not (Rich et al., 1977). Reduction of the U(VI) can be effected by reducing agents such as sulfides, iron(II), and organic matter. More information regarding the uranium in its ores can be found in many texts and monographs (IAEA 1974, 1976, 1993).

Oxidation of the tetravalent uranium in both the uraninite and pitchblende leads to the introduction of the hexavalent uranium in the form of the uranyl ion, UO_2^{2+}, which can form the basis of an extensive chemistry of its own. In addition to the formation of the ion itself from oxidation, it reacts with many other cations and anions to form other complex molecules. This is attested to by the existence of mixed cation uranyl minerals containing a wide variety of metals ions resulting in minerals such as saleeite ($Mg(UO_2)_2(PO_4)_2 \cdot 10(H_2O)$), dewindtite ($Pb_3[H(UO_2)_3O_2(PO_4)_2]_2 \cdot 12(H_2O)$), and torbernite ($Cu(UO_2)_2(PO_4)_2 \cdot 8\text{-}12(H_2O)$). Uranium(VI) forms "simple" compounds such as carbonates (i.e., UO_2CO_3). Additionally, the uranyl ion can undergo reactions with many organic compounds to form coordination complexes, including the uranyl ion in conjunction with ammonia, urea, and many oxygen donor molecules. The chemistry of the uranium and its oxides is treated in a number of standard works (Burns and Finch, 1999; Katz et al., 1986; Raran, 1994).

Research Needs and Opportunities

The Environmental Management Science Program (EMSP) should support near-term (1–5-year) research to help ensure safety of the depleted uranium hexafluoride during storage, transportation, and conversion. The EMSP should also support longer-term research that might lead to new, beneficial uses for uranium or that would provide a scientific basis for selecting a disposal option.

Near-Term Research for Managing and Converting the DUF_6

Even though DU is only slightly radioactive, its concentration in large masses in the DUF_6 cylinders produces radiation doses to workers in their vicinity. The radiation levels measured on the outside surface of cylinders are typically about 2 to 3 millirem per hour (mrem/hr) decreasing to about 1 mrem/hr at a distance of 1 foot.[2] Although the historical dose to cylinder yard workers has been less than 200 mrem/yr, the committee was told during its visit that the cylinder yard is one of the more significant sources of worker doses at the Oak Ridge Reservation (Philpot, 2002).

During its visit, the committee observed that the way the cylinders are stacked restricts the workspace between cylinders and in some cases precludes workers from being able to examine the entire outer surface of each cylinder. Nor is it possible to confidently move and hoist all cylinders because corrosion may have weakened some to the point that they could be damaged by the available handling techniques and equipment (see Figure 6.4). As time passes, more cylinders will fall into this category. If a cylinder is breached the release of UF_6 and its reaction products (e.g., HF), could have serious consequences.

There is need and opportunity for near-term (1–5-year) research that will support DOE's plans for converting its DUF_6 to oxide. Particularly

---

[2] http://web.ead.anl.gov/uranium/guide/health.

Figure 6.4 Currently available equipment may not be adequate to safely lift severely corroded cylinders. Source: http://web.ead.anl.gov/uranium/guide/.

with emphasis on robotics or remotely operated methods that enhance worker safety, research could lead to new techniques and equipment to

- assess the integrity of DUF_6 cylinders whose exterior surface is partially blocked, and which cannot be handled with confidence;
- extract the DUF_6 from cylinders in the storage yard that cannot be moved to the processing facility, or whose integrity cannot be ascertained; and
- provide field deployable methods to measure contaminants in the DUF_6, which might be important for operator safety or to meet regulatory requirements for transporting or converting the material. For example, some cylinders are contaminated with technetium from recycled uranium.

Longer-Term Research for Reuse or Disposal

WHO has compiled a list of the research needed to better assess chemical and radiological health risks from exposure to uranium compounds.[3] The committee believes that this research will assist DOE in its future decisions for reusing or disposing of its DU:

- Neurotoxicity: Other heavy metals (e.g., lead and mercury) are known neurotoxins, but only a few studies have been conducted on uranium. Studies are needed to determine if DU is neurotoxic. Reproductive and developmental effects have been reported in single animal studies but no studies have been conducted to determine if they can be confirmed or that they occur in humans.
- Hematological effects: Uranium distribution within bone is thought to be such that irradiation of bone marrow and blood-forming cells are limited due to the short range of alpha particles emitted during decay. Research is needed to determine if this view is correct.
- Genotoxicity: Some in vitro studies suggest genotoxic[4] effects occur via the binding of uranium compounds to DNA. Research is needed to determine if uranium is genotoxic by this or other mechanisms.

---

[3] See http://www.who.int/environmental_information/radiation/depleted_uranium.htm.

[4] Genotoxic refers to materials that are capable of causing damage to genetic material (DNA). DNA damage does not lead inevitably to the creation of cancerous cells, but potentially such damage can lead to the formation of a malignancy.

There are also opportunities to extend current knowledge in the following areas:

- Understanding of the extent, reversibility, and possible existence of thresholds for kidney damage in people exposed to DU. Important information could come from studies of populations exposed to naturally elevated concentrations of uranium in drinking water.
- Better assessments of impacts of exposure of children. This is particularly important given their unique exposure scenarios such as geophagia and hand-to-mouth activities.
- Validation of transfer coefficients for uranium compounds entering the food chain, for example, from soil ingested by livestock during grazing and then to humans.

Investigations are needed on the chemical and physical form, physiological behavior, leaching, and subsequent environmental cycling of specific forms of uranium from various industrial and military sources (e.g., depleted uranium alloys, phosphate by-products). Particular attention should be paid to how the bulk of DU might eventually be disposed.

Aside from the possible presence of contaminants in some of the DU from recycled uranium, the isotope enrichment process leaves a material that initially has a lower radioactivity than natural uranium. Not only U-235 but most of the uranium decay chain isotopes (e.g., radium, radon) are removed. Modeling the long-term behavior of DU should include the fact that these daughter isotopes will gradually reappear over time.

Reuse

Uranium compounds have been used as colorants in ceramic glazes (e.g., “Fiestaware”). DU has been proposed as a diluent for some spent nuclear fuel, to ensure that stored fuel elements do not achieve criticality, and for excess HEU nuclear fuels to render them less attractive as potential weapons material. Uranium silicides are potential fuels, and research into such alloy fuels would be facilitated by the availability of DU for processing and radiation effects stability studies. DU is a candidate fertile material for future breeder reactors. Steel/UO_2 cermets, which were developed as test reactor fuels, are a candidate material for use as gamma-shielding repository containers for spent fuel (Forsberg and Haire, 2002; Haire, 2002). Given its range of chemical valence states and redox potentials, uranium could have important applications in catalysis, optics, and electronics.

Disposal

Returning the DU, as oxide, to former uranium mines is attractive because it does not foreclose recovery and reuse options. The goal of research directed toward this option should be to understand and hence avoid mechanisms that might lead to rapid migration of uranium from the disposal facility. Research opportunities include the study of the interaction chemistry of uranium and its oxides with reactants that might be found under environmental conditions in a mine or near-surface repository—reactants such as water and carbon dioxide. This can include the identification of reactions to form new uranium phases, in both the U(IV) and U(VI) oxidation states. Some possibilities might be mixed carbonate oxide phases resulting from CO_2 reactions, uranium oxide/metal ion reactions involving other metal ions that are present in a uranium mine setting, and the study of interfacial reactions involving aqueous solutions of metal ions with uranium and its oxides.

These studies can include electronic and magnetic state data, since uranium—under normal environmental conditions—exhibits a $5f^2$ electronic configuration for U(IV) and a $5f^0$ electronic configuration for U(VI). Reactions of importance also encompass chemisorption, reduction/oxidation related to metal ion/uranium ion couplets, and the study of some of these reactions as a function of segregation and diffusion in rock matrices. Saturated aqueous solution studies of uranium oxides with mineral phases that might exist in uranium mine environments are relevant. Oxidation of UO_2 under different chemical conditions is of interest, especially for input into models. Research approaches might ideally combine spectroscopic techniques with microscopy in order to study the chemistry of the uranium system with respect to chemical changes, species, and physical as well as chemical phases.

7

The Higher Actinides

The United States' massive program to produce nuclear materials, which lasted for over 50 years, was halted in 1992. With the closure of its production reactors and separations facilities the Department of Energy (DOE) no longer has the capability for large-scale production of transuranic isotopes (see Chapter 2 and Appendix A). DOE's inventory, however, contains substantial amounts of the higher actinides,[1] many of which were produced in special campaigns that involved multiple irradiation and separation steps. Currently there is little or no use foreseen for these isotopes and for the most part they are considered a liability by DOE's Office of Environmental Management (EM), which is charged with cleaning up the former production sites. In addition, the facilities for handling and storing these isotopes are being closed as part of the cleanup. Consequently, EM plans to dispose of many unique materials as waste (e.g., by adding them to high-level tank waste). This route will foreclose all other options and risks future regret of an irrevocable action.

Starting with uranium, the higher actinide isotopes are built up by successive, non-fission-producing neutron capture in the parent nucleus followed by β-particle emission (see Figure 7.1). Because neutron capture usually splits the nucleus (fission), the yield of the higher actinides decreases rapidly as atomic number and mass increase. As noted in Chapter 3, DOE produced about 100 metric tons of plutonium-239. By contrast, kilogram quantities of americium-243 and gram quantities of californium-252 are considered large amounts. Known uses for some of the higher actinides will consume only a fraction of the available inventory.

---

[1] Defined for the purpose of this report as isotopes having an atomic number of 93 (Np) or greater.

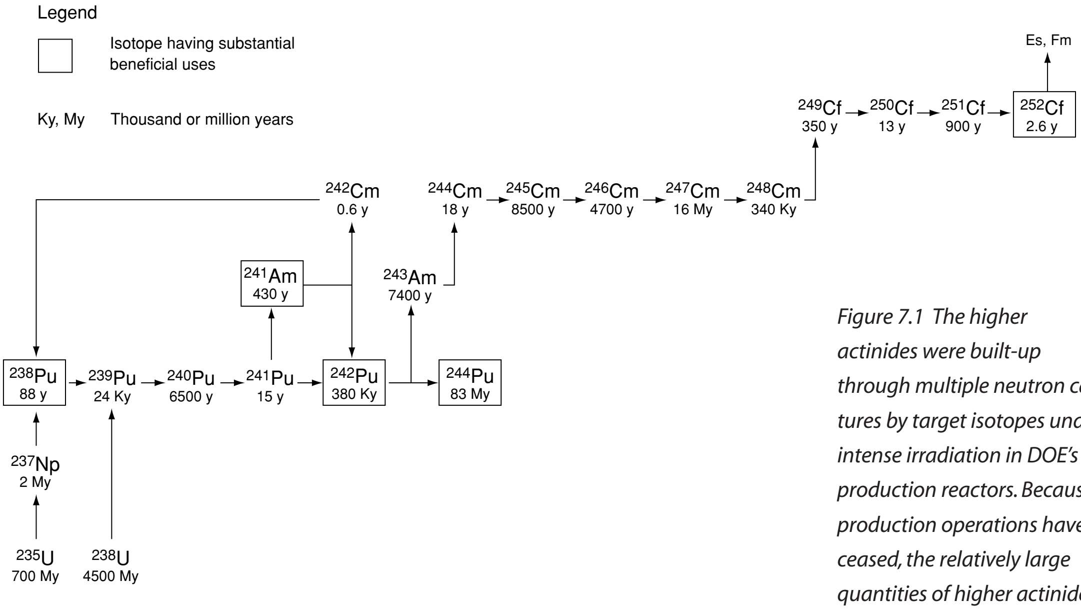


Figure 7.1 The higher actinides were built-up through multiple neutron captures by target isotopes under intense irradiation in DOE's production reactors. Because production operations have ceased, the relatively large quantities of higher actinide isotopes in DOE's inventory cannot be replaced. Source: Croff et al., 2002.

As is the case with DOE's other legacy nuclear materials discussed in this report, there has been comparatively little research toward beneficial uses of the higher actinides. The committee regards this as a knowledge gap rather than an intrinsic shortcoming of the materials.

The committee recommends that the EMSP should support research to preserve and stabilize the inventory of higher actinide isotopes, identify beneficial new uses, and develop a better understanding of their radiological and chemical health effects.

There is need and opportunity for the Office of Science to play a lead role in ensuring that the United States maintains both the facilities and the skilled personnel necessary to preserve and to conduct research with DOE's unique separated isotopes. EMSP-funded research can play an important role in this effort.

Neptunium-237 and Plutonium-238

Plutonium-238 for use in thermoelectric power systems in the space program is produced by reactor irradiation of Np-237. During reactor irradiation, neutron capture by Np-237 produces Np-238, which beta-decays with a half-life of 2.1 days to Pu-238. Neptunium-237, a fissile isotope, is obtained by reprocessing reactor fuels.

About a third of the thermal neutron captures by U-235 lead to U-236 rather than fission of the U-235. During production operations enriched uranium fuel at Savannah River Site (SRS) was recovered, converted to metal at Oak Ridge, and recycled back to SRS fuel elements to build the U-236 in successive irradiation cycles. As U-236 increases, further neutron capture gives U-237, which decays to Np-237 by beta emission with a short half-life of 6.75 days. A lesser production route has been by (n, 2n) reactions on U-238 by the relatively small flux of fast neutrons in the production reactors. Neptunium-237 has a half-life of about 2 million years and decays by alpha emission.

About 100 to 200 kg of Np-237 will be needed to support the space program's requirement for Pu-238 for the next 35 years (Croff et al., 2002). This supply is assured if DOE is able to convert the main Np-237 inventory, stored in solution at SRS, to a solid for shipment to Oak Ridge and if adequate reactor and processing infrastructure remains at Oak Ridge. Oak Ridge researchers (Collins et al., 2002) have also proposed to produce Pu-238 from Am-241 that will be recovered from weapons-grade plutonium to be converted into mixed oxide fuel at SRS (see Chapter 3). This would also require maintaining reactor and processing infrastructure at Oak Ridge.

Mossbauer spectroscopy is an especially useful method for basic research on the chemical properties of Np-237, such as electronic and magnetic bonding data. Many studies have been reported in the chemical literature, and a substantial experimental database exists for Np-237 Mossbauer parameters as a function of oxidation state and chemical species (Greenwood and Gibb, 1971; Long, 1984). There are research opportunities to use this technique for fundamental studies that may lead to more efficient separation processes or potentially new uses for neptunium.

Americium-243 and Curium-244

Americium-243 and curium-244 were produced in extensive campaigns at SRS and Oak Ridge from 1961 to 1975. Cm-244 was expected to become a substitute for Pu-238 in thermoelectric power units for space missions. Cm-244, with an 18-year half-life, provides 2.8 watts (thermal) per gram versus 0.57 watts per gram for Pu-238 with its 86-year half-life. A second objective was to use the Cm-244 as a target for building up to Cf-252, which can be used as a point source of neutrons, as discussed in the next section. Am-243 and Cm-244 are essential feedstocks for production of the heavier curium isotopes, californium, and higher isotopes.

The major campaign to produce americium and curium began at SRS by long irradiations of about 100 kg of Pu-239 to enhance the Pu-

240 content. This was followed by fabrication of plutonium-aluminum alloy targets and re-irradiations for two and a half years in a production reactor specially reconfigured to produce an extremely high neutron flux.[2] Overall the program encompassed ten different processing campaigns with up to three target irradiation histories per campaign.

This effort produced about 10 kg of Am-243 and 2.6 kg of Cm-244. The expected markets for Cm-244 and Cf-252 did not materialize, and these materials were left in solution in the SRS F-Canyon. At the time the committee completed its work, in fall 2002, DOE had closed F-Canyon and had firm plans to dispose of its Am/Cm solution into the million-gallon-capacity high-level waste tanks at SRS. An additional 750 grams of curium are in unreprocessed targets (Mark 18A targets), which were also left as a legacy of the campaign. Besides these legacy materials, about 800 grams of curium are held at Oak Ridge National Laboratory (ORNL) and SRS for use by the DOE Office of Science to produce higher actinides. Disposing of the SRS F-Canyon solution will result in the loss of most of the U.S. feedstock for producing higher actinides.

Plutonium-244

Plutonium-244 is the product of years of intense neutron irradiation of plutonium in nuclear reactors.[3] It is used in very small quantities in isotope dilution mass spectrometry to precisely measure the isotopic composition of plutonium being safeguarded by U.S. and international organizations such as the International Atomic Energy Agency. Only gram quantities were made, and the current supply of separated Pu-244 is essentially exhausted. The aforementioned Mark 18A targets, however, contain about 20 grams of unseparated Pu-244. This Pu-244 is now viewed as a national resource, mainly because of new concerns over weapons proliferation, and it is likely that the legacy Mark 18As will be retained, shipped to ORNL, and reprocessed as needed (Croff et al., 2002).

---

[2] The highest flux was $6x10^{15}$ neutrons per square centimeter per second, and flux greater than $1x10^{15}$ over a reactor volume 6 feet high and 7 feet in diameter. This is about three times higher flux over a much greater volume than is available today for isotope production, for example, in the High Flux Isotope Reactor at Oak Ridge.

[3] Traces of primordial Pu-244 have been detected on Earth (Hoffman et al., 1971).

Californium-252

Californium-252 was first identified in the debris from thermonuclear test explosions. Currently ORNL produces about 0.25 grams of this isotope per year from SRS feedstock. The only other production is at Dmitrovgrad, Russia, which has a capacity of about one-tenth that of ORNL.

The primary use of Cf-252 is in brachytherapy for the treatment of certain cancers.[4] In addition, during the past 45 years, Cf-252 has been used successfully for neutron radiography of objects ranging from flowers to entire aircraft; startup sources for nuclear reactors; and elemental analysis of coal, nuclear fuel, explosives, and the human body, among other uses (Osborne-Lee and Alexander, 1995). As noted earlier, the large demand for Cf-252 envisioned in the mid-1970s did not materialize. Recently, in treating muscular sarcomas; skin, head and neck tumors; and gynecological tumors, brachytherapy has shown high cure rates and lower recurrence rates compared to other treatment options such as surgery (Fontanesi et al., 1999; Maruyama, 1984 and Maruyama et al., 1986; Rivard et al., 1999; Taeev et al., 2001). Medical applications for radioisotopes are expanding rapidly (see Sidebar 7.1), and there appears to be a good possibility that there will be resurgence in the use of Cf-252.

Challenges and Opportunities for Preserving the Inventory

The higher actinides in the DOE inventory represent material that may be useful in its present form, may be suitable for target material, or may be essential for research into developing new materials. However, the inventory is part the legacy of former U.S. weapons production, which is generally considered to be undesirable, expensive, and hazardous. The committee concluded that there are three principal challenges to preserving the inventory:

---

[4] Neutron brachytherapy is a treatment using neutron beams produced by a Cf-252 source. The term "brachy" (from the Greek *brachios*) literally means "close by" and is used to refer to radiation therapy applied over a short distance, usually with a high degree of localization and precision. In brachytherapy, a beam of neutrons from a Cf-252 source provides a high radiation dose to the tumor while minimizing the dose to surrounding healthy tissues.

SIDEBAR 7.1 MEDICAL USES OF ISOTOPES

There are many radioisotopes in use for medical purposes. The DOE Office of Isotopes for Medicine and Science Programs has published a list of major isotopes and their uses (http://www.ne.doe.gov/isotope/isotop.html#top). New developments continue to arise, such as radioactively tagged monoclonal antibody treatments for leukemia using bismuth-213 being studied at New York City's Sloan Kettering Cancer Center, at ORNL, and at the National Cancer Institute (DOE, 2001c; Wu, 1997).

Isotope demand versus availability has been addressed previously by an Institute of Medicine report (IOM, 1995). More recently, the demand and availability of medical isotopes was discussed in a joint meeting between the DOE and the National Cancer Institute. The DOE supply of isotopes (Lowe, 2002) in particular was discussed, including near-term and long-term plans. Five facilities are available for isotope production. Two accelerators will continue to operate (Brookhaven and Los Alamos); and three reactors will continue production (Oak Ridge, Idaho, and Sandia). The Isotope Production Facility at Los Alamos is expected to come on line in 2004. A new production cyclotron will come on line in 2008, possibly at Brookhaven National Laboratory. The acquisition of a dedicated, single-mission, isotope production and processing facility that will be fully operational by 2010 is planned to include a cyclotron and a reactor both dedicated to isotope production.

Projections of demand for medical isotopes forecast a sharp rise for both therapeutic and diagnostic isotope products, as shown in Figure 7.2 (Burns, 2002). While there is some debate about specific nuclides and rates of growth (Wagner and Reba, 1999), a recent expert panel reported general agreement on the following trends: (http://www.ne.doe.gov/nerac/finalisotopereport.pdf):

- **increased growth in isotope use,**
- **shortages of some major nuclides,**
- **lack of a reliable supply of research isotopes produced at a reasonable cost,**
- **deteriorating DOE infrastructure, and**
- **an overdependence on non-U.S. radionuclide production.**

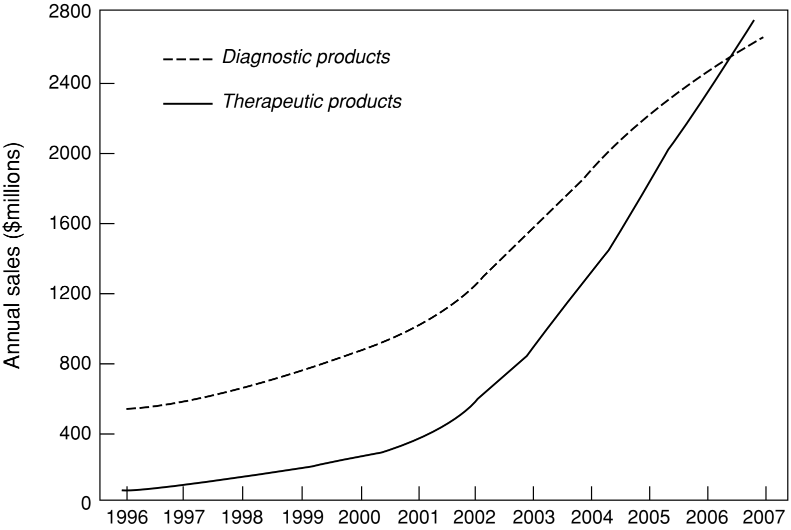


Figure 7.2 The medical use of radioisotopes is expected to continue growing. In addition to Cf-252, other higher actinides may have beneficial applications in medicine. Source: Burns, 2002.

- Facilities capable of handling or storing the materials are being closed.
- Few new nuclear scientists are being trained.
- Accumulated knowledge, both documentation and personal expertise, is being lost.

The committee believes that the Office of Science's mission, which includes stewardship of human resources, essential scientific disciplines, institutions, and scientific facilities, and fostering dissemination of information (Dehmer, 1998), is well suited to meet these challenges.

The Office of Science has an opportunity to lead other DOE offices and industrial partners in establishing a "center of excellence" to ensure that the United States has a continuing capability to handle and store large inventories of higher actinides for research, beneficial use, or as feedstock. A precedent for such a center was the ORNL Heavy Isotope Working Group, which was funded by DOE-Oak Ridge and EM's Office of Nuclear Material and Spent Fuel (EM-21) in FY 2001. The center's scope need not be limited to the higher actinides, rather its function could include lighter radioisotopes and cross-cut research for characterizing and stabilizing the plutonium, depleted uranium, and spent nuclear fuel described earlier in this report.

A similar center of excellence concept was offered by a previous National Academies' committee, which recommended that EMSP consider establishing field sites for conducting subsurface contamination research to "attract new researchers to the program, encourage both formal and informal multidisciplinary collaborations among researchers, and facilitate the transfer of research results into application" (NRC, 2000, p. 8). A field research site is being established at the Idaho National Engineering and Environmental Laboratory.[5]

The committee also noted the tremendous amount of research, design, test, and evaluation work that was conducted at the DOE production sites. Much of the research and development needed for managing DOE's nuclear materials has already been done at the production sites. Reports have been either archived or classified to the extent that many are neither searchable nor easily retrievable, and hence likely to be lost. Another function of the center would be to bring this documentation into the modern world of information management.

---

[5] See http://subsurface.inel.gov/information/sgl.asp.

Radiation Effects on Organisms

Most of the motivation for EM to dispose of the higher actinides is the perception that they are a hazard to people or the environment.[6] However, the scientific basis for determining the biological and environmental effects of the higher actinides and consequently their risks and disposal requirements is limited (see, for example, Sontag, 1983). Understanding their risks is also essential for developing beneficial future uses.

The risks presented by radionuclides have largely been dictated by their potential impact on people. For the past 50 years, what and how much can be released has been derived from acceptable doses to affected persons. First, an acceptable dose limit (rem or Sv) to humans was specified. Then, an allowable annual intake of a particular radionuclide to achieve that limit was calculated (Ci or Bq per year). All the routes (pathways) the radionuclide(s) could take from the source to the receptor were identified, and the fractional transport through each environmental medium was estimated. Finally, the allowable contamination level was back calculated—based on the exposure scenarios, length of time from release to receptor, and other factors. While this methodology has been an integral part of managing radioactive materials for several decades, there is substantial room for improvement in the process. Importantly, any reduction in conservatism in any of the assumptions could lead to a substantive impact on how radionuclides are handled and ultimately disposed.

All of our current models and standards are based on protecting humans. The general adequacy of the models has been shown with regard to food chain transport to people. Consequences to nonhuman biota have largely been ignored. It has been assumed (ICRP, 1991) that protection of the general public automatically confers an adequate level of protection for the environment. Recent developments have shown that models used to assess the potential impacts on humans from radionuclide releases to the environment may not adequately predict the potential impacts to biota (DOE, 2002b; IAEA, 1992, 2002a, 2002b; Stone, 2002). According to the IAEA (2002b, p. 6),

> For biota, the concept of dose is not yet fully developed. In principle, the calculation of absorbed dose, including the distribution of absorbed dose between different organs or tissues, is feasible.

---

[6] DOE justifies its decisions based on cost. Costs of the EM cleanup program are largely determined by the belief that DOE's former production sites present major health and safety risks.

However, in practice, the ability to do so is limited by the lack of data on the uptake of radionuclides by the wide range of organisms, which are of interest, and on the distribution of radionuclides between different internal organs and tissues. Currently, most assessments of radiation doses to biota have estimated absorbed doses averaged throughout the whole organism. There is no consensus on the use of radiation weighting factors (values ranging from 5 to 40 for alpha radiation have been suggested, and used in dose assessments). An international consensus on this issue would be valuable.

Specific areas of research recommended by the IAEA include:

- long-term radioecological studies (many of these have been abandoned or mothballed at a number of major DOE facilities—some of these studies had been in place for 30-40 years and could be resumed);
- biotic uptake factors by species, environment, and nuclide;
- dosimetry calculations for reference organisms;
- biological elimination functions and rates by species;
- environmental transfer rates;
- dose-effect studies;
- radiation weighting factors; and
- development of environmental models that focus on pathways other than those leading to humans.

Stabilization and Reuse

EMSP research toward assuring safe, inexpensive, long-term storage of the higher actinides can play an important role in preserving them for beneficial future uses. Medical uses of Cf-252 appear to be increasing, as discussed in the previous section. Medical uses of other actinides, including the trans-californium isotopes, may be discovered. There are opportunities for research on both the radiological and the chemical effects of the higher actinides in biological systems. This research can lead to better understanding of both the harmful effects these materials might have in the environment and their potentially beneficial uses in medicine.

There are opportunities for research toward new uses for the higher actinides in industry. In Russia, californium has been used mainly in industrial applications rather than in medicine. Uses have included continuous neutron activation analyses in mining operations to sort and grade various ores, reducing production costs. One variation was on ships recovering undersea manganese or other nodules, to monitor intake and discard low-grade material continuously instead of hauling it to shore.

Neutron moderation measurements were used to detect water condensed or frozen inside airframe walls, which adversely affects aircraft performance—particularly in Siberia. A recent National Research Council report (NRC, 2002) on retrieving buried waste noted new technologies being developed by the U.S. Department of Defense that utilize Cf-252 in land mine detection.

Cm-244 with its half-life of 18 years constitutes a relatively rapidly decreasing stockpile of feed for higher elements. Continuing irradiation to yield Cm-246 and Cm-248 with half-lives of 4,730 years and 340,000 years would give essentially a non-decaying stockpile. There are research opportunities in basic physics, chemistry, and materials science to determine the feasibility of this transformation, which would be essential if significant amounts of curium are to be preserved for more than about 50 years.

The high flux reactor at the Institute at Dimitrovgrad has a modified neutron energy spectrum that enhances the capture to fission ratio markedly, giving improved yields of heavy nuclides. Use of this reactor to produce the more stable curium isotopes may have practical benefits as well as encouraging more international collaboration in research on the higher actinides.

Because the higher actinides are stored as relatively pure, separated materials—rather than in the highly heterogeneous mixtures that are characteristic of wastes—they are good candidates for long-term storage or disposal in specifically tailored ceramic forms. There are research opportunities to develop ceramics with lattice configurations and radiation response that will ensure that they are permanently immobilized. See Chapter 3 for a detailed discussion of ceramics for plutonium.

A variety of analytical techniques are available to study the basic chemistry of the higher actinides and for developing immobilization matrices. One of the most broadly applicable is X-ray photoelectron spectroscopy (XPS). Survey scans using the technique can be used to determine the identity and composition of the material with respect to the elements present, while high-resolution spectra can be obtained to get a detailed understanding of the chemistry of the materials, chemical alterations, and impurities. Detailed chemical information can be obtained, including oxidation states, chemical functional groups in which the elements are contained, and chemical species of the elements. The reaction chemistry involving the materials can be followed, especially reactions with water, gases, and other fluids. XPS has been used extensively to study uranium, thorium, plutonium, and other actinides with respect to both their electronic and magnetic properties and their reaction chemistries.

Auger electron spectroscopy (AES) also has been used widely in materials science studies, including its extensive use in corrosion studies. In addition to giving basic spectral data, it can be used as a microprobe-based characterization technique to image and analyze the surface of materials. Auger spectra may be produced by both electron and X-ray beams. With the latter source also producing XPS spectra, one can obtain combined XPS-AES spectra and take advantage of additional spectral parameters on chemical species that cannot be obtained by either XPS or AES alone. The element of imaging adds the possibility of obtaining morphological data concerning interfacial reactions on material surfaces.

Both XPS and AES are highly useful in looking at the detailed chemistry of actinides in crystalline matrices. The two techniques can be coupled with microscopy to look at virtually any materials chemistry problem that these forms pose. Phenomena such as elemental segregation, redox chemistry, grain boundary problems, dissolution, and chemisorption can be addressed with these two techniques (Briggs and Seah, 1990).

8

Conclusions

The overarching theme that guided the committee's deliberations throughout this study is that scientific research begun now can inform the Department of Energy's (DOE's) future decisions for disposition of surplus nuclear materials. DOE should avoid decisions today that foreclose future options. Basic physical and chemical principles guarantee that there will be no simple, shortcut ways to replace the currently available quantities of nuclear materials that resulted from 50 years of intense effort in the United States' massive nuclear complex during the Cold War.

The large-inventory nuclear materials dealt with in this report (plutonium, spent nuclear fuels, depleted uranium, cesium-137 and strontium-90 capsules, and higher actinides) have only been available for a few decades. The next few decades may bring unforeseen beneficial uses so that these materials are recognized as valuable and irreplaceable resources. The committee concluded that the Environmental Management Science Program (EMSP) and the Office of Science have a unique opportunity to provide leadership and, by partnering with other DOE offices and private industry, financial resources to

- ensure that the inventory is stabilized for safe, economical, long-term storage;
- maintain the intellectual and facility infrastructure for world-class research and development activities with the inventoried materials; and
- seek beneficial uses for these materials.

Research Priorities

The EMSP's congressional charter calls for long-term, path-breaking research. In addition, opportunities for research that provides a high potential payoff in addressing urgent, near-term needs may arise. As a practical matter, the EMSP may well encounter a range of research opportunities that span short- and long-term needs. In addition, EMSP projects may well include a technology component necessary for transitioning basic research to application (DOE, 2000a).

The research recommendations in Chapters 3-7 suggest that the greatest opportunities for research that might provide shorter-term (1-5 year) payoffs are generally in the area of stabilizing the inventory for storage. Specific examples include:

- Plutonium: stabilize for up to 50 years of storage according to the DOE 3013 Standard.
- DOE spent nuclear fuel: arrest cladding degradation and prepare for several decades of storage before eventual disposal in a geological repository.
- Cs-137 and Sr-90 capsules: understand fundamental mechanisms that lead to swelling and potential breaching of containment.
- Depleted uranium: provide new methods to ensure safe removal of DUF_6 from deteriorated containers to support DOE's plan for converting the DUF_6 to a stable oxide.
- Higher actinides: develop stable forms to allow their removal from closure sites, shipment, and storage at sites with a continuing nuclear materials mission.

Begun now, longer-term research would feed a continuously growing body of scientific information to support decision making and have the potential of eventually providing scientific breakthroughs. Longer-term research should be directed toward beneficial new uses for DOE's nuclear materials or their disposal. Examples include:

- Plutonium: maximize the portion of the scrap inventory that can be converted to MOX fuel.
- DOE spent nuclear fuel: ensure that it meets waste acceptance criteria for disposal with a minimum amount of treatment.
- Cs-137 and Sr-90: develop stable, non-dispersible matrices for storage to decay (several hundred years) or disposal.
- Depleted uranium and heavy actinides: strive to develop beneficial uses along with further understanding health effects that might suggest or preclude some uses; provide a scientific basis for disposing the DU oxide and heavy actinides.

Crosscutting Opportunities

This report is the last in a series of five National Academies' studies requested by the EMSP to assist in providing an agenda for research to support and enhance DOE's site cleanup program (NRC, 2000, 2001a, 2001b, 2002). The previous reports dealt with environmental contamination and waste issues. Most of the excess nuclear materials that are the subject of this report have not been declared as waste, and according to its statement of task the committee emphasized research directed toward preserving and reusing the materials. Nevertheless, there is a broad consistency among the recommendations in all five studies. Three areas stand out as offering opportunities for the EMSP to support scientific research that crosscuts most of DOE's cleanup challenges. For DOE's excess nuclear materials, wastes, and contaminated media crosscutting topics for research include:

- characterization of their fundamental chemical, physical, and biological properties;
- treatment to ensure their near- and long-term stability, including understanding the fundamental parameters that affect stability; and
- assessment of their actual health or environmental risks.

By focusing its limited funds in these crosscutting areas, and leveraging its limited funding by cooperative research with other DOE offices or the private sector, the EMSP is most likely to achieve the scientific breakthroughs intended by its congressional charter.

References

Abu-Qare, A. W., and M. B. Abou-Donia. 2002. Depleted uranium—the growing concern. Journal of Applied Toxicology 22(3): 149-152.

Anderson, S., S. Nesbit, J. Gilreath, and T. Laubham. 2002. Mixed Oxide Fuel Effects on the Integrity of the McGuire and Catawba Reactor Vessels. Fifth Topical Meeting on Spent Nuclear Fuel and Fissile Materials Management, Charleston, South Carolina, September 17-20. La Grange Park, Illinois: American Nuclear Society.

Bailey, G., E. Bluhm, J. Lyman, R. Mason, M. Paffett, G. Polanskey, G.D. Roberson, M. Sherman, K. Veirs, and L. Worl. 2000. Gas Generation from Actinide Oxide Materials. LA-13781-MS. Los Alamos National Laboratory: Department of Energy.

Berg, J., D. Harradine, D. Hill, J. McFarlane, D. Padilla, F. C. Prenger, D. K. Veirs, and L. Worl. 2002. Gas Generation Testing Over Plutonium Oxides in the 94-1 Shelf-Life Surveillance Program. Fifth Topical Meeting on Spent Nuclear Fuel and Fissile Materials Management, Charleston, South Carolina, September 17-20. La Grange Park, Illinois: American Nuclear Society.

Briggs, D., and M.P. Seah, Eds. 1990. Practical Surface Analysis: Auger and X-Ray Photoelectron Spectroscopy. New York: John Wiley & Sons Ltd.

BSC (Bechtel SAIC Company). 2001. Performance Assessment of U.S. Department of Energy Spent Fuels in Support of Site Recommendation. CAL-WIS-PA-000002 revision 00. Las Vegas, Nevada: BSC.

Burns, M. 2002. The New Era of Therapeutic Radiopharmaceuticals. Joint Department of Energy and National Cancer Institute Meeting, Los Angeles, California, June 17. http://www.ne.doe.gov/isotope/conference/NewEra.pdf (accessed September 7, 2002).

Burns, P.C., and R. Finch, Eds. 1999. Uranium: Mineralogy, Geochemistry, and the Environment. Reviews in Mineralogy and Geochemistry. Volume 38. Washington, D.C.: Mineralogical Society of America.

Chambers, B., and T. Kiess. 2002. USDOE Nuclear Material Disposal Orphans. Waste Management-02. Tucson, Arizona: WM Symposia, Inc.

Clark, D.E., W.B. White, and A.J. Machiels, Eds. 1986. Nuclear Waste Management II: Advances in Ceramics. Volume 20. Columbus, Ohio: American Ceramic Society.

Clemen, R.T. 1996. Making Hard Decisions: An Introduction to Decision Analysis, 2nd edition. Belmont, California: Wadsworth Publishing Company.

Collins, E.D., G.E. Michaels, C.W. Alexander, J.E. Bigelow, and R.M. Wham. 2002. A New Potential for Recovery and Use of Americium-241. Fifth Topical Meeting on Spent Nuclear Fuel and Fissile Materials Management, Charleston, South Carolina, September 17-20. La Grange Park, Illinois: American Nuclear Society.

Croff, A.G., R.M. Canon, C.W. Alexander, E.D. Collins, and F. Peretz. 2002. Production, Uses, Supply, and Demand of Heavy Actinide Isotopes. Fifth Topical Meeting on Spent Nuclear Fuel and Fissile Materials Management, Charleston, South Carolina, September 17-20. La Grange Park, Illinois: American Nuclear Society.

Dehmer, P.M. 1998. Assessing the value of research at the Department of Energy: A perspective from the Office of Basic Energy Sciences. In: Assessing the Value of Research in the Chemical Sciences: Report of a Workshop. Washington, D.C.: National Academy Press.

DNFSB (Defense Nuclear Facilities Safety Board). 1994. Improved Schedule for Remediation in the Defense Nuclear Facilities Complex. Washington, D.C.: Defense Nuclear Facilities Safety Board. May.

DNFSB. 1996. Trip Report—Safety of Cesium and Strontium Capsules at Hanford. Washington, D.C.: Defense Nuclear Facilities Safety Board. June.

DNFSB. 2002. Savannah River Site Canyon Utilization. DNFSB/TECH-32. Washington, D.C.: Defense Nuclear Facilities Safety Board. March.

DOE (U.S. Department of Energy). 1994a. Assessment of Plutonium Storage Safety Issues at Department of Energy Facilities. DOE/DP-0123T. Washington, D.C.: Office of Defense Programs.

DOE. 1994b. High-Level Waste Borosilicate Glass—A Compendium of Corrosion Characteristics. DOE-EM-0177. Washington, D.C.: Office of Environmental Management.

DOE. 1996a. Closing the Circle on the Splitting of the Atom: The Environmental Legacy of Nuclear Weapons Production in the United States and What the Department of Energy Is Doing About It. DOE/EM-0266. Washington, D.C.: Office of Environmental Management.

DOE. 1996b. Final Environmental Impact Statement for the Tank Waste Remediation System. Richland, Washington: U.S. Department of Energy and Washington State Department of Ecology. April.

DOE. 1998a. Accelerating Cleanup: Paths to Closure. DOE/EM-0362. Washington, D.C.: Office of Environmental Management.

DOE. 1998b. Guidelines for Risk-Based Prioritization of DOE Activities. DOE-DP-STD-3023-98. Washington, D.C.: Office of Defense Programs.

DOE. 2000a. Environmental Management Science Program. Annual Report. FY 2000. Office of Science and Technology. DOE/EM-0569. Washington, D.C.: Office of Environmental Management.

DOE. 2000b. Stabilization, Packaging, and Storage of Plutonium-Bearing Materials. DOE-STD-3013-2000. Washington, D.C.: National Nuclear Security Administration.

DOE. 2000c. A Strategic Approach to Integrating the Long-Term Management of Nuclear Materials: DOE's Integrated Nuclear Materials Management Plan. Washington, D.C.: Department of Energy.

DOE. 2001a. Summary Data on the Radioactive Waste, Spent Nuclear Fuel, and Contaminated Media Managed by the U.S. Department of Energy. Washington, D.C.: Office of Environmental Management.

DOE. 2001b. Fiscal Years 2001-2005 Multiyear Program Plan: Office of Science and Technology. DOE/ID-10840. Washington, D.C.: Office of Environmental Management.

DOE. 2001c. Report to Congress on the Extraction of Medical Isotopes from Uranium-233. Washington, D.C.: Office of Nuclear Energy, Science and Technology, and Office of Isotopes for Medicine and Science. March.

DOE. 2002a. A Review of the Environmental Management Program. Presented to the Assistant Secretary for Environmental Management by the Top-to-Bottom Review Team. Washington, D.C.: Office of Environmental Management. February.

DOE. 2002b. A Graded Approach to Evaluating Radiation Doses to Aquatic and Terrestrial Biota: Final Technical Standard. DOE-STD-1153-2002. Washington, D.C.: U.S. Department of Energy.

DOE. 2002c. Amendment to the Record of Decision on the Waste Isolation Pilot Plant: Disposal Phase Supplemental Environmental Impact Statement. Washington, D.C.: Office of Environmental Management. November.

DOE. 2002d. Supplemental Analysis for the Disposal of Certain Rocky Flats Plutonium-Bearing Materials at the Waste Isolation Pilot Plant. DOE/EIS-0026-SA-3. Washington, D.C.: Office of Environmental Management.

DOE. 2002e. Department of Energy Selects Uranium Disposition Services for Uranium Hexafluoride Conversion Plants in Ohio and Kentucky. DOE News. Washington, D.C.: Office of Public Affairs. August 29.

DOE. 2002f. Contact-Handled Transuranic Waste Acceptance Criteria for the Waste Isolation Pilot Plant. DOE/WIPP-02-3122 revision 0.1. Carlsbad Field Office: Department of Energy.

DOE. 2002g. Performance Management Plan for the Accelerated Cleanup of the Hanford Site. DOE/RL-2002-47 revision D. Richland, Washington: Department of Energy.

DOE. 2003. Advanced Fuel Cycle Initiative: The Future Path for Advanced Spent Fuel Treatment and Transmutation Research. Report to Congress. Washington, D.C.: Office of Nuclear Energy, Science, and Technology. January.

Domenici, P.V. 2002. Letter to The Honorable Spencer Abraham regarding disposal of Rocky Flats plutonium waste in the WIPP. Washington, D.C.: U.S. Senate. February 5.

Duffey, J., and R. Livingston. 2002. Gas Generation Testing of Plutonium Dioxide. Fifth Topical Meeting on Spent Nuclear Fuel and Fissile Materials Management, Charleston, South Carolina, September 17-20. La Grange Park, Illinois: American Nuclear Society.

Duguid, J., R. K. Senger, and J. Leem. 2002. Performance Assessment of a Post Closure Pyrophoric Event. Fifth Topical Meeting on Spent Nuclear Fuel and Fissile Materials Management, Charleston, South Carolina, September 17-20. La Grange Park, Illinois: American Nuclear Society.

Fontanesi, J.N., P.J. Chuba, M. Rivard, R. Benda, F.E. Rector, and D.L. Farmer. 1999. Pediatric Brachytherapy: The Barbara Ann Karmanos Cancer Institute experience using Californium-252. Journal of Brachytherapy International 15:37-41.

Forsberg, C.W., and M. J. Haire. 2002. Depleted Uranium Dioxide-Steel Cermets for Spent Nuclear Fuel Multipurpose Casks. Fifth Topical Meeting on Spent Nuclear Fuel and Fissile Materials Management, Charleston, South Carolina, September 17-20. La Grange Park, Illinois: American Nuclear Society.

Freshley, M.D. 1973. UO_2-PuO_2: A demonstrated Fuel for Plutonium Utilization in Thermal Reactors. Nuclear Technology 18:141-170.

Greenspan, E., N.Z. Cho, S.G. Hong, T.H. Kim, and W. E. Kastenberg. 1998. Multi-Recycling of Spent Fuel with Low Proliferation Risk. Proc. 9th International Conference on Emerging Nuclear Energy Systems, ICENES ‘98. The Netherlands: Nuclear Research and Consultancy Group. June.

Greenwood, N.N., and T.C. Gibb. 1971. Mossbauer Spectroscopy. London: Chapman & Hall.

Haire, M.J. 2002. The U.S. Depleted Uranium Uses Research and Development Project. Presented to the National Research Council's Committee on Improving the Scientific Basis for Managing Nuclear Materials and Spent Nuclear Fuel through the Environmental Management Science Program. Oak Ridge, Tennessee. March 6.

Herald. 2002. DOE draft plan to speed Hanford cleanup unveiled. Richland, Washington: Tri-City Herald. May 2.

Hobbs, L.W., A.N. Sreeram, B. Berger, and C.E. Jesurum. 1996. Structural freedom, topological disorder, and the irradiation-induced amorphization of ceramics structures. Nuclear Instruments and Methods in Physics Research B 116:17-25.

Hobbs, L.W., C.E. Jesurum, and B. Berger. 1999. Rigidity constraints in the amorphization of singly- and multiply-polytopic structures. In: Rigidity Theory and Applications, P.M. Duxbury and M.F. Thorpe, Eds. New York: Plenum Press. Pp. 191-216.

Hoffman D.C., F.O. Lawrence, J.L. Mewherter, and F.M. Rourke. 1971. Detection of Plutonium-244 in Nature. Nature 234:132-134. November 19, 1971.

IAEA (International Atomic Energy Agency). 1974. Formation of Uranium Ore Deposits. Proceedings of a Symposium on the Formation of Uranium Ore Deposits held May 6-10. Vienna: International Atomic Energy Agency.

IAEA. 1976. Uranium Ore Processing. Proceedings of an Advisory Group Meeting held November 24-26, 1975. Vienna: International Atomic Energy Agency.

IAEA. 1992. Effects of Ionizing Radiation on Plants and Animals at Levels Implied by Current Radiation Protection Standards. Technical Reports Series No. 332. Vienna: International Atomic Energy Agency.

IAEA. 1993. Analytical Techniques in Uranium Exploration and Ore Processing. Technical Report Series No. 341. Vienna: International Atomic Energy Agency.

IAEA. 2002a. Ethical Considerations in Protecting the Environment from the Effects of Ionizing Radiation. IAEA-TECDOC-1270. Vienna: International Atomic Energy Agency.

IAEA. 2002b. Specialists Meeting on Protection of the Environment from the Effects of Ionizing Radiation: International Perspectives. IAEA-723-J9-SP-114.3. Vienna: International Atomic Energy Agency.

IAEA 2003. International Conference on the Security of Radioactive Sources held March 10-13. Vienna: International Atomic Energy Agency.

ICRP (International Commission on Radiological Protection). 1991. Recommendations of the International Commission on Radiation Protection (1990). Publication 60. Oxford and New York: Pergamon Press.

INEEL (Idaho National Engineering and Environmental Laboratory). 2001. An Integrated Roadmap for the Programmatic Resolution of Gas Generation Issues Containing Radioactive Waste Materials. Final Report Issued by National Transportation Integration and Planning Program. Idaho Falls, Idaho: Bechtel BWXT. April.

IOM (Institute of Medicine). 1995. Isotopes for Medicine and the Life Sciences. Washington, D.C.: National Academy Press.

Johnson, J., and E. Brabazon. 2002. Design of the MOX Fuel Fabrication Facility. Fifth Topical Meeting on Spent Nuclear Fuel and Fissile Materials Management, Charleston, South Carolina, September 17-20. La Grange Park, Illinois: American Nuclear Society.

Katz, J.J., G.T. Seaborg, and L.R. Morss, Eds. 1986. Chemistry of the Actinide Elements, 2nd edition. Dordrecht, Netherlands: Kluwer Academic Publishers.

Keeney R.L., and T.L. McDaniels. 2001. A framework to guide thinking and analysis regarding climate change policies. Risk Analysis 21(6): 989-1000.

Kolman, D. 2001. A review of the potential environmentally assisted failure mechanisms of austenitic stainless steel storage containers housing stabilized radioactive compounds. Corrosion Science. 43:99-125.

Leeds, E.J. 2000. Comments on DUF_6 Materials Use Roadmap. Letter to the Depleted Uranium Hexafluoride Management Program (NE-30) of the U.S. Department of Energy. Washington, D.C.: U.S. Nuclear Regulatory Commission. October 18.

Long, G.J. 1984. Mossbauer Spectroscopy Applied to Inorganic Chemistry. Modern Inorganic Chemistry. New York: Plenum Publishing.

Long, M.E. 2002. America's Nuclear Waste. National Geographic Magazine. July, 2002. Pp. 8-33.

Lowe, O. 2002. DOE Isotope Supply 2002-2010. Joint Department of Energy and National Cancer Institute Meeting, Los Angeles, California. June 17.

Maruyama, Y., Ed. 1984. Cf-252 Neutron Brachytherapy: An Advance for Bulky Localized Cancer Therapy. New York: Taylor & Francis.

Maruyama, Y., J.L. Reach, and J.M. Feda, Eds. 1986. Californium-252 Brachytherapy and Fast Neutron Beam Therapy. New York: Taylor & Francis.

McDiarmid, M. A., K. Squibb, S. Engelhardt, M. Oliver, P. Gucer, P.D. Wilson, R. Kane, M. Kabat, B. Kaup, L. Anderson, D. Hoover, L. Brown, and D. Jacobson-Kram. 2001. Surveillance of depleted-uranium-exposed Gulf War veterans: Health effects observed in an enlarged "friendly fire" cohort. Journal of Occupational and Environmental Medicine 43(12): 991-1000.

McKenney, D.E., and T.L. Walton. 2001. Waste Management Project, Science and Technology Plan. Richland, Washington: Fluor Hanford. September.

Miller, A.C., J. Xu, M. Stewart, P.G.S. Prasanna, and N. Page. 2002. Potential late health effects of depleted uranium and tungsten used in armor-piercing munitions: Comparison of neoplastic transformation and genotoxicity with the known carcinogen nickel. Military Medicine 167(2nd Suppl.): 120-122.

NAS (National Academy of Sciences). 1994. Management and Disposition of Excess Weapons Plutonium. Washington, D.C.: National Academy Press.

NAS. 1995. Management and Disposition of Excess Weapons Plutonium: Reactor Related Options. Washington, D.C.: National Academy Press.

NAS. 2000. The Spent-Fuel Standard for Disposition of Excess Weapon Plutonium: Application to Current DOE Options. Washington, D.C.: National Academy Press.

Naugle, K. 2002. Cycle Design Work for Transition to Partial MOX Fuel Cores. Fifth Topical Meeting on Spent Nuclear Fuel and Fissile Materials Management, Charleston, South Carolina, September 17-20. La Grange Park, Illinois: American Nuclear Society.

NEA (Nuclear Energy Agency). 2001. Management of Depleted Uranium. A joint report of the Organisation for Economic Co-Operation and Development Nuclear Energy Agency (NEA-OECD) and the International Atomic Energy Agency. Paris: NEA.

NRC (National Research Council). 1996. The Hanford Waste Tanks: Environmental Impacts and Policy Choices. Washington, D.C.: National Academy Press.

NRC. 1997a. Building an Effective Environmental Management Science Program: Final Assessment. Washington, D.C.: National Academy Press.

NRC. 1997b. Glass as a Waste Form and Vitrification Technology: Summary of an International Workshop. Washington, D.C.: National Academy Press.

NRC. 1998. Research Reactor Aluminum Spent Fuel: Treatment Options for Disposal. Washington, D.C.: National Academy Press.

NRC. 2000. Research Needs in Subsurface Science: U.S. Department of Energy's Environmental Science Program. Washington, D.C.: National Academy Press.

NRC. 2001a. Research Needs for High-Level Waste Stored in Tanks and Bins at U.S. Department of Energy Sites. Washington, D.C.: National Academy Press.

NRC. 2001b. Research Opportunities for Deactivating and Decommissioning Department of Energy Facilities. Washington, D.C.: National Academy Press.

NRC. 2002. Research Opportunities for Managing the Department of Energy's Transuranic and Mixed Wastes. Washington, D.C.: National Academy Press.

NRC. 2003. One Step at a Time: The Staged Development of Geologic Repositories for High-Level Radioactive Waste. Washington, D.C.: National Academy Press.

Osborne-Lee, I.W., and C.W. Alexander. 1995. Californium-252: A Remarkably Versatile Radioisotope. ORNL/TM-12706. Oak Ridge, Tennessee: Oak Ridge National Laboratory.

Patrinos, A. 2002. Environmental Remediation Sciences Update. Presented to the National Research Council's Board on Radioactive Waste Management. Washington, D.C. December 12.

Pedersen, R.C. 2002. Advanced Test Reactor Irradiation of MOX with Comparisons to PWR Conditions. Fifth Topical Meeting on Spent Nuclear Fuel and Fissile Materials Management, Charleston, South Carolina, September 17-20. La Grange Park, Illinois: American Nuclear Society.

Philpot, H. 2002. East Tennessee Technology Park UF_6 Cylinder Management Tour. Presented to the National Research Council's Committee on Improving the Scientific Basis for Managing Nuclear Materials and Spent Nuclear Fuel through the Environmental Management Science Program. Oak Ridge, Tennessee. March 6.

Raran, V. 1994. Uranium(VI)-Oxygen Chemistry: Uranyl Hydroxo Complexes, Uranates, Oxides. Palm Harbor, Florida: Hadronic Press.

Rich, R.A., H.D. Holland, and U. Petersen. 1977. Hydrothermal Uranium Deposits. Amsterdam: Elsevier Scientific Publishing.

Rivard, M.J., J.G. Wierzbicki, F. Van den Heuvel, R.C. Martin, and R.R. McMahon. 1999. Clinical brachytherapy with neutron emitting Cf-252 sources and adherence to AAPM TG-43 dosimetry protocol. Medical Physics 26(1): 87-96.

ROO (DOE Richland Operations Office). 2002. Waste Encapsulation and Storage Facility (WESF) Fact Sheet. REG-0275. Richland, Washington: Department of Energy.

Sickafus, K.E., L. Minervini, R.W. Grimes, J.A. Valdez, M. Ishamaru, F. Li, K.J. McClellan, and T. Hartmann. 2000. Radiation tolerance of complex oxides. Science 289:748-51.

Siskin, E. 2002. Revised U.S. Plutonium Disposition Strategy. Presented to the National Research Council's Board on Radioactive Waste Management. Washington, D.C. July 30.

Sontag, W. 1983. The early distribution of plutonium-239, americium-241 and uranium-233 in the soft tissues and skeleton of old rats. A comparative study. Human Toxicology 2(1): 91-100.

SROO (DOE Savannah River Operations Office). 2002. Hanford Nuclear Materials Disposition Planning. Memorandum from Michael G. O'Rear, Director, National Materials Management Division. Aiken, South Carolina: Department of Energy. April.

Stone, R. 2002. Radioecology's coming of age—Or its last gasp. Science 297:1801. September 13.

Taeev, T., B. Ptaekova, V. Strnad, H. Koukalova, V. Kry Tof, and O. Ra Ovska. 2001. Importance of Californium-252 for a Better Radio-Curability of Carcinoma of the Cervix Uteri: Long-Time Treatment Results Presented. Klinick Onkologie 14:59-64.

Tri Party. 2002. Letter of Intent to Accelerate Hanford Cleanup. State of Washington, U.S. Environmental Protection Agency, U.S. Department of Energy, March 5. Available at http://www.em.doe.gov/Letter_of_IntentWA_508.pdf (accessed March 26, 2003).

Tseng, J.C. 2001. Nuclear Material and Spent Fuel Environmental Management. Presented to the National Research Council's Committee on Improving the Scientific Basis for Managing Nuclear Materials and Spent Nuclear Fuel through the Environmental Management Science Program. Washington, D.C. October 20.

USNRC (U.S. Nuclear Regulatory Commission). 1989. Recent Safety-Related Incidents at Large Irradiators. Information Notice No. 89-82. Washington, D.C.: U.S. Nuclear Regulatory Commission. December 7.

Venetz, T. 2002. Personal communication to Mark Paffett. Richland Washington: Fluor-Daniel Richland Operations. May.

Wagner, H. N., and R.C. Reba. 1999. Expert Panel Forecast of Future Demand for Medical Isotopes. U.S. Department of Energy. March. http://www.ne.doe.gov/nerac/isotopedemand.pdf (accessed September 7, 2002).

Wang, S.X., B.D. Begg, L.M. Wang, R.C. Ewing, W.J. Weber, and K.V.G. Kutty. 1999. Radiation stability of gadolinium zirconate: A waste form for plutonium disposition. Journal of Materials Research 14:4470.

Weber, W.J., R.C. Ewing, C.R.A. Catlow, T. Diaz de la Rubia, L.W. Hobbs, C. Kinoshita, H. Matzke, A.T. Motta, M. Nastasi, E.H.K. Salje, E.R. Vance, and S.J. Zinkle. 1998. Radiation effects in crystalline ceramics for the immobilization of high-level nuclear waste and plutonium. Journal of Materials Research 13:1434-1484.

Whicker, F. W., and V. Schultz. 1982. Radioecology: Nuclear energy and the environment. Volume 1. Boca Raton, Florida: CRC Press.

WHO (World Health Organization). 2001. Depleted Uranium Sources, Exposure and Health Effects. WHO/SDE/PHE/01. Geneva: World Health Organization. April.

WSRC (Westinghouse Savannah River Company). 2002. Nuclear Materials Disposition Pathways for the Hanford Site. WSRC-TR-2002-00178. Aiken, South Carolina: WSRC.

Wu, C. 1997. New Drug Zaps Cancer Cells with Radiation. Science News Online. February 27. http://www.sciencenews.org/sn_arc97/2_22_97/fob2.htm (accessed September 7, 2002).

Appendixes

Appendix A

Nuclear Materials Production in the DOE Complex

Nuclear weapons production in the United States was a complex series of integrated activities at multiple sites across the country. To provide a context for the nuclear materials and spent fuel challenges and research opportunities described in this report, these activities can be grouped into seven major processes:

- mining, milling, and refining of uranium;
- isotope separation of uranium, lithium, boron, and heavy water;
- fuel and target fabrication for production reactors;
- reactor operations to irradiate fuel and targets to produce nuclear materials;
- chemical separations of plutonium, uranium, and tritium from irradiated fuel and target elements;
- component fabrication of both nuclear and nonnuclear components; and
- weapon operations, including assembly, maintenance, modification, and dismantlement of nuclear weapons.

Uranium Processing and Enrichment

Uranium production began with mining and milling to extract uranium ore from the Earth and chemically processing it to prepare uranium concentrate (U_3O_8), sometimes called uranium octaoxide or yellowcake. Because natural uranium consists mainly of the mass 238 isotope (U-238) and only about 0.7 percent of the fissile isotope, U-235, the next step was to concentrate (enrich) the U-235 content in a portion of the uranium. The process began with natural uranium and resulted in enriched uranium and depleted uranium. The first U.S.

uranium enrichment facilities were located in Oak Ridge, Tennessee. Additional enrichment plants were later built in Piketon, Ohio, and Paducah, Kentucky.

Highly enriched uranium (HEU) contains 20 weight percent or more of U-235; it was fashioned into weapon components and also used as reactor fuel. Low enriched uranium (LEU), which contains less than 20 weight percent of U-235, and natural uranium were used as reactor fuel for plutonium production. Depleted uranium was used in weapon components and for Pu-239 production. All the uranium enriched during the Manhattan Project was HEU for weapon components. However, as early as 1950, LEU was used for reactor fuel.

Uranium enrichment has resulted in the accumulation of about 700,000 metric tons of depleted uranium hexafluoride (DUF_6), most of which was stored in large carbon steel cylinders at the enrichment facilities. The DUF_6 comprises the largest quantity of separated material in the DOE complex. Research opportunities that might lead to improved options for management, reuse, or disposal of this material are discussed in Chapter 6.

Nuclear Fuel and Reactor Operations

The focus of the Department of Energy's (DOE's) nuclear materials production activities was to produce plutonium for nuclear weapons.[1] Enriched uranium served as fuel in production reactors, and excess neutrons from the nuclear chain reaction bred Pu-239 and other isotopes in "targets" made of U-238. Fuel and target fabrication consisted of the foundry and machine shop operations required to convert uranium feed material, principally metal, into fuel and target elements. Some later production reactors used separate fuel and target elements, while early production reactors used the same elements for both fuel and targets. Uranium ingots were extruded, rolled, drawn, swaged, straightened, and outgassed to produce rods and plates. The rods were machined, ground, cleaned, coated, clad, and assembled into finished fuel.

Reactor fuel and target fabrication was initially carried out by private contractors and at the Hanford, Washington, and the Savannah River, South Carolina, production reactor sites. Within a decade, government-owned plants in Fernald, Ohio, and Weldon Spring, Missouri, took over part of this mission, supplying the fuel manufacturing plants at Hanford and the Savannah River Site (SRS). At SRS, fuel rods were made by extrusion of an alloy of aluminum and HEU to form thin-walled, aluminum-clad fuel tubes.

---

[1] Tritium is not dealt with in this report.

Reactor operations include loading and removal of fuel and target elements, reactor maintenance, and the operation of the reactor itself. Early experimental reactors were built at Oak Ridge, Hanford, and in the Chicago, Illinois, area. Nine full-scale production reactors were located at Hanford, and five others were built at the SRS.

Reactor operations created essentially all the nuclear materials used in the DOE complex. Except for a few special cases, such as research reactor fuel, the highly radioactive spent fuel and targets were reprocessed to recover plutonium, uranium, and other isotopes and to separate waste materials. However, when the United States stopped its plutonium production in 1992, some spent fuels, including targets, were left unreprocessed. Currently, DOE's inventory of spent nuclear fuels (SNF) amounts to about 2,500 metric tons of heavy metal (U and Pu), most of which are stored at Hanford, SRS, Oak Ridge, and the Idaho National Engineering and Environmental Laboratory (INEEL). Chapter 4 describes research needs and opportunities for improving DOE's ability to manage and dispose of its SNF in view of their potential radiation and security risks.

Chemical Separations

Chemical separation involved dissolving SNF and targets and isolating and concentrating the plutonium, uranium, and other nuclear materials they contained. Three basic chemical separation processes were used on a production scale in the United States: bismuth phosphate, reduction oxidation, and plutonium uranium extraction (PUREX). Chemical separation plants were located at Hanford, SRS (see Sidebar A.1), and INEEL.

Chemical separation of spent fuel and target elements produced large volumes of highly radioactive waste (high-level waste), and large quantities of low-level radioactive wastewater, solid low-level waste, and mixed low-level waste. Dealing with these waste materials is a central part of the DOE Office of Environmental Management's cleanup mission. Previous National Academies' reports have provided advice to the Environmental Management Science Program on research to improve management of these wastes. Separated nuclear materials from reprocessing that are dealt with in this report include plutonium (Chapter 3), cesium and strontium (Chapter 5), and special isotopes (Chapter 7).

Weapons Activities

Weapons operations include the assembly, maintenance, and dismantlement of nuclear weapons. Weapons operations were chiefly done at the Pantex Plant near Amarillo, Texas, the Iowa Army Ordnance Plant in Burlington, Technical Area 2 of Sandia National Laboratories, New Mexico, and the Clarksville, Tennessee, and Medina, Texas, modification centers.

Assembly is the process of joining together separately manufactured components and major parts into complete, functional, and certified nuclear warheads for delivery to the Department of Defense. Maintenance includes the modification and upkeep of a nuclear weapon during its life cycle. Dismantlement involves the reduction of retired warheads to a nonfunctional state and the disposition of their component parts. The dismantlement process yields parts containing special nuclear materials, high explosives, hazardous materials, and other components with hazardous and nonhazardous properties. Some parts are returned to the facility where they were originally produced. Other parts are maintained in storage (e.g., plutonium pits) or are dispositioned on site. With respect to the excess plutonium, a major step toward disposition will be conversion to mixed oxide fuel for commercial power reactors at a new facility to be built at SRS (see Chapter 3).

SIDEBAR A.1 NUCLEAR MATERIALS PRODUCTION AT THE SAVANNAH RIVER SITE

The primary processing facilities at SRS are the F- and H-Canyons and B Lines (finishing facilities), with F-Canyon starting into production in late 1954 and H-Canyon starting in mid-1955. The two canyons were similar when first constructed but were modified over the years to provide separate capabilities, though many operations can be done in either, but at different rates. Originally, both utilized the PUREX solvent extraction process to separate plutonium from irradiated natural uranium. The original B-Lines were based on the plutonium peroxide, plutonium tetrafluoride, calcium reduction route to metal. The installation also incorporated recovery facilities for slag and crucible, out of specification material, and other residues, because an original goal was that no backlog of recoverable plutonium was to be accumulated. From 1957 to 1959, F-Canyon was shut down for the installation of higher-capacity equipment for solvent extraction and a new plutonium finishing line based on a plutonium fluoride precipitation route to metal. Later, more recovery capacity was added. Meanwhile, H-Canyon continued in plutonium production. During this period, reactor operation changed to driver elements of HEU and targets of DU metal for plutonium production and of lithium-aluminum alloy for tritium production.

Operation of F-Canyon restarted in 1959, and H-Canyon was shut down and modified to maintain nuclear safety while processing HEU driver elements. Changes included dissolver inserts to provide safe geometry, lowered concentration of the tributylphosphate extractant, and instruments to monitor and control concentrations of the uranium in the liquid phases. Only a few months were required for production in H-Canyon to resume.

A number of functions and capabilities were added to the separations facilities for special programs. Recovery of Np-237, fabrication of reactor targets, and separation and recovery of neptunium and

Pu-238 from the targets were provided by canyon installations and finishing facilities in H-Canyon. Special dissolver inserts allowed wide varieties of fuels to be processed, including enriched fuels being returned from domestic and foreign research reactors. An electrolytic dissolver was utilized for some stainless steel- and zirconium-clad fuels. The ability to remotely rearrange flow routes and equipment was utilized in processing irradiated thorium to recover U-233.

Many separate campaigns were involved in the program to produce transplutonium elements such as curium-244 and californium, which required repeated recoveries, target fabrications, and reirradiations of plutonium fractions. For the californium program, a special section was carved out of the far end of the F-Canyon for the installation of the Multi-Purpose Processing Facility. This consisted of a group of small racks containing capabilities for dissolving, chromatic ion exchange, precipitation, and calcining operations. A legacy from that program is the Am/Cm solution discussed in Chapter 7. Special plutonium irradiation campaigns were made to produce various isotopic compositions of plutonium that would be approached in a plutonium breeder economy where plutonium would be recycled back into fuel. These materials went to tests to determine reactor neutronic characteristics at different stages of plutonium recycle operation. The H-Canyon B-Line can process Np-237, Pu-238, and Pu-239. The F-Canyon B-Line recovery can process slags and crucibles from metal production and miscellaneous scrap.

As of the summer of 2002, the last plutonium metal has been produced in F B-Line, the liquid system has been flushed, and preparations are under way to put F-Canyon on standby. The F B-Line dry mechanical line will be used to calcine plutonium returns to meet specifications on moisture and volatile materials, utilizing new high-temperature furnaces that can reach the specified firing temperature of 1000 °C. Products are to be packaged in both inner and outer containers to meet the 3013 Standard for storage containers (see Chapter 3).

H-Canyon will continue to process the backlog of aluminum-clad enriched fuels for some years and has the capability to process some plutonium materials. The present primary route for disposition of enriched uranium fuels is to process them for purification and blend the uranium down to nominally 4 percent enrichment for transfer to the Tennessee Valley Authority and to reactor fuel. Other enriched uranium fuels would be sent to a geological repository. A variety of plutonium scrap and mixed plutonium-uranium material will be treated in H B-Line with some plutonium going to mixed oxide fuel, some to waste and then the Defense Waste Processing Facility, and some to storage to await decisions on eventual disposition.

Appendix B

Biographical Sketches of Committee Members

WM. HOWARD ARNOLD (NAE) (Chair) retired as general manager of the Advanced Energy Systems Division of Westinghouse Electric Company. His primary areas of expertise include nuclear power, fuel, and waste disposal. He has designed nuclear reactor cores for civilian power reactors, for space power and propulsion, and for production of nuclear materials. He has managed multidisciplinary groups of engineers and scientists working in reactor core design and led work that promoted the use of centrifuge technology in uranium enrichment. As vice president of Westinghouse Hanford Company, he was responsible for engineering, development, and project management at the Hanford Site from 1986 to 1989. Recently Dr. Arnold has been involved in an advisory capacity in the cleanup of Department of Energy (DOE) nuclear weapons material production sites, especially in the vitrification plant at the Savannah River Site. He received his A.B. in 1951 from Cornell University, his M.A. in 1953, and his Ph.D. in physics in 1955, both from Princeton University.

GREGORY B. COTTEN is an assistant professor at the U.S. Naval Academy, Annapolis, Maryland. His expertise is in chemical separations and waste processing. He has seven years of experience as a project engineer and manager at the Idaho National Engineering and Environmental Laboratory. He also served for six years as a submarine officer during which he qualified as engineering officer of the watch on pressurized water reactor plants. He is a member of the American Nuclear Society and the National Society of Professional Engineers. He received the Idaho Society of Professional Engineers Outstanding Young Engineer of the Year award in 2000. Dr. Cotten is author or co-author of about 25 publications. He received his Ph.D. (2000), and M.E.

(1997), degrees in chemical engineering from the University of Idaho. His B.S. degree (1987) is from the U.S. Naval Academy.

KATHRYN B. HIGLEY is an associate professor of radiation health physics in the Department of Nuclear Engineering at Oregon State University. Her expertise is in assessing the effects of radiation on humans and in the environment, nuclear emergency response planning, and environmental regulations. She has three years experience in environmental radiation monitoring at Trojan Nuclear Power Plant and 14 years with Battelle Pacific Northwest Laboratories as an environmental health physicist. She has been a consultant to DOE’s Office of Environment, Safety and Health, Pacific Northwest National Laboratory, and Argonne National Laboratory. She was president of the Health Physics Society environmental section (1998-1999), and has served on National Council on Radiation Protection and Measurements subcommittees and as a member of the American Board on Health Physics panel of examiners. She is a certified health physicist, member of the Health Physics Society, Society of Environmental Toxicology and Chemistry, and BIOMOVs II (Biospheric Model Validation Study). Dr. Higley received her Ph.D. (1994) and M.S. (1992) degrees in radiological health sciences from Colorado State University, and her B.A. degree in radiochemistry (1978) from Reed College.

LINN W. HOBBS is professor of materials science and professor of nuclear engineering at Massachusetts Institute of Technology, where he was the inaugural holder of the John F. Elliott Chair of Materials. His expertise is in characterization, using electron microscopy, diffraction methods, and computer simulation, of atomic and extended defect structures and microstructures that are introduced within inorganic materials by radiation or chemically driven compositional change. He has authored over 200 journal articles and eleven book chapters, and authored or edited seven books. Dr. Hobbs has a longstanding research interest in the use of glass and crystalline wasteforms for storage of radioactive nuclear waste, served on the NRC Committee on Long-Term Research Needs for Radioactive High-Level Waste at DOE Sites, and is the principal organizer for the Engineering Conferences International (ECI) international conference on Alternative Nuclear Wasteforms to be held in January 2004. He is a fellow and director of the American Ceramic Society, a former president of the Microscopy Society of America, and a former councilor of the Materials Research Society. He chaired the British Marshall Scholarships northeast regional selection committee for 13 years, and serves on the Truman Scholarships selection committee. Dr. Hobbs received his B.Sc. in materials science from Northwestern University and his D.Phil. in science of materials from

Oxford University. He was made an officer of the Order of the British Empire in 2001.

DONALD A. ORTH is an independent consultant following his retirement as a departmental fellow, E.I. du Pont de Nemours and Co., and consulting scientist, Westinghouse Savannah River Co. During his 40-year career he did basic work on development, design, and operation of processes and facilities for nuclear materials production. This included production of plutonium, californium, other transplutonium elements, and uranium and thorium isotopes. After retirement, he has participated in site visits to review Russian transplutonium element production and utilization programs as well as annual reviews of the Argonne National Laboratory program on electromechanical treatment of spent nuclear fuels. He received the Glenn T. Seaborg Award in actinide separations in 1990. Upon his retirement in 1992, the Westinghouse Savannah River Company created the Donald A. Orth award for technical excellence, which is presented annually. Dr. Orth received his B.S degree in 1948 and his Ph.D. degree in 1951, both in chemistry, from the University of California, Berkeley.

IRVIN OSBORNE-LEE is an associate professor and head of the Department of Chemical Engineering at Prairie View A&M University. Previously he spent 13 years in the Chemical Technology Division of Oak Ridge National Laboratory. His expertise is in developing disposition pathways and treatment methods for problematic wastes. He has authored or co-authored about 50 papers in this area. He is also committed to positively impacting society through academic enterprise: educating and empowering students, motivating and inspiring faculty, and building key research programs. His honors and awards include the 2001 Appreciation Award of the National Society of Black Engineers and the Service to Society Award of the American Institute of Chemical Engineers (AIChE), in which he has held a number of positions. Dr. Osborne-Lee is a member of the AIChE, National Organization for the Professional Advancement of Black Chemists and Chemical Engineers, Sigma Xi, and the National Council of Black Engineers and Scientists. He received his Ph.D., M.E., and B.S. degrees from the University of Texas, Austin, in 1985, 1983, and 1979, respectively, all in chemical engineering.

MARK T. PAFFETT is a New Mexico native who has been at Los Alamos National Laboratory (LANL) since 1983. He began as a postdoctoral fellow in E and CHM divisions working under the supervision of Dr. Charles T. Campbell. He matriculated from the University of New Mexico in 1978 with a B.S. in chemistry (with honors) and Ph.D. in chemistry from the California Institute of Technology in 1983. In the ensuing years his research endeavors have included fuel cell electrocatalysis, heterogeneous catalysis, surface science, and analytical chemistry. Dr. Paffett is the surface science team leader in Chemistry-Spectroscopy and Inorganic Chemistry and project leader for the 94-1 Program. His current research thrusts include fundamental and programmatic research on surface radiolytic and thermal processes over surfaces of importance to LANL. These studies have included in situ studies using IR spectroscopy, gas-phase catalytic measurements of actinide materials, a variety of thermal experimental surface science studies, and chemical kinetic modeling studies of chemistry initiated by surface radiolytic processes. In addition, he has built and maintains extensive analytical surface capabilities that include XPS, SIMS, SNMS, AES, and IR techniques (many of these coupled via load locks to high pressure or specialized microreactors). He has published over 85 refereed publications and has been cited over 1,900 times.

DALE L. PERRY is a senior scientist in chemistry at the Lawrence Berkeley National Laboratory, Berkeley, California. His expertise is in the chemistry and synthesis of inorganic systems, including those of actinides, lanthanides, and transition metal ions. He has served on several DOE panels related to instrumentation needs in actinides and heavy metals in the environment, including the Fernald Uranium Production Facility Panel for Closure Characterization. He has acted as adviser to DOE and other federal agencies in chemistry and materials chemistry as they relate to national security issues. He is the author and co-author of over 300 refereed journal publications, book chapters, and presentations. He is a member of the American Chemical Society, Materials Research Society, the Society for Applied Spectroscopy, and the Society for the Advancement of Chicanos and Native Americans in Science. In 2002, he received a DOE Outstanding Mentor Award for his work related to his involving students in research. He is a Fellow of the Royal Society of Chemistry (London) and a Fellow of the American Association for the Advancement of Science. He was the 1997 National Chairman for the Industrial & Engineering Chemistry Division of the American Chemical Society. Dr. Perry received his Ph.D. in inorganic chemistry from the University of Houston in 1974.

PER F. PETERSON is professor and chair of nuclear engineering at the University of California, Berkeley. His expertise is in energy and environmental systems, including passive reactor safety systems, inertial fusion energy, and nuclear materials management. He worked at Bechtel on high-level radioactive waste processing from 1982 to 1985. He was a fellow at the Tokyo Institute of Technology from 1989 to 1990 and a National Science Foundation Presidential Young Investigator from 1990 to 1995. He is past chairman of the Thermal Hydraulics Division of the American Nuclear Society (1996-1997) and has served as editor for three journals. He is the author of over 60 archival journal articles and over 60 conference publications. He received his Ph.D. (1988) and M.S. (1986) degrees in mechanical engineering the University of California, Berkeley. His B.S. degree (1982), also in mechanical engineering, is from the University of Nevada, Reno.

STEVEN M. THORNBERG is a staff member at Sandia National Laboratories. His expertise is in developing analytical standards and quality assurance methods for nuclear weapons stockpile surveillance. Recently his work has focused on the analysis of gases produced by materials aging and radiolysis as well as pressure, vacuum, and leak rate measurement of containers for storing materials and fissile isotopes in the DOE weapons stockpile stewardship program. He has participated in several DOE audits, including a sitewide Environmental Health and Safety "tiger team" audit in 1989-1990, the "red team" audit of the Hanford tank farms in 1992, and audits of analytical standards laboratories at several sites. Dr. Thornberg received his Ph.D. in analytical chemistry from the University of New Mexico, Albuquerque, in 1984, and his B.A., magna cum laude, from Western State College, Gunnison, Colorado, in 1980.

ROBERT W. YOUNGBLOOD is vice president and chief technical officer of the Nuclear Systems Analysis Division of Information Systems Laboratories, Inc. His areas of expertise are probabilistic risk analysis and reliability analysis. Dr. Youngblood has worked extensively with the Nuclear Regulatory Commission, including assignments as principal investigator in developing an approach to performance-based regulation and as task leader in assessing options for improved regulation of by-product materials systems. For DOE, he served on a panel to recommend responses to concerns raised by the Defense Nuclear Facility Safety Board, and he was principal investigator for a project to develop and prescribe methods and assumptions for analysis of radiological accidents at a high-level waste facility. He has developed software to support risk analysis and applications of risk analysis. Dr. Youngblood received his Ph.D. (1976) and M.S. (1970) degrees from the State University of New York at Stony Brook, and his B.A. degree from Reed College (1968), all in physics.

Appendix C

Presentations to the Committee

Washington, D.C., October 24-25, 2001

Overview of the Office of Science and Technology's Role in the Environmental Management Cleanup Mission, Teresa Fryberger, Department of Energy (DOE)

Charter and Description of the EM Science Program, Roland Hirsch and Mark Gilbertson, DOE

Nuclear Material Program Review, John Tseng, DOE

Spent Nuclear Fuel Program Review, John Tseng, DOE

Role of the Nuclear Materials Focus Area (NMFA), Stan Wolf, DOE

Depleted Uranium Program Review, Kevin Shaw, DOE

Statement of Task and Needs of the Sponsor, Gerald Boyd and Mark Gilbertson, DOE

Santa Fe and Albuquerque, New Mexico, January 14-15, 2002

EM Program Oversight for Pu Operations, R. Erickson, Los Alamos National Laboratory (LANL)

NMFA Work at LANL and Emerging Needs, J. Boak, LANL

MOX Fuel from Excess Pu, K. Abney, LANL

Aries Program for Stabilizing Pu, C. James, LANL

NDA Assay Techniques, N. Enslinn, LANL

Roundtable Discussions with LANL Research Personnel

Roundtable Discussions with Sandia National Laboratories Personnel

Oak Ridge, Tennessee, March 6-7, 2002

Oak Ridge Nuclear Materials Overview, Allen Croff, Oak Ridge National Laboratory (ORNL)

Depleted Uranium (DU), J. Haire and R. Hightower, ORNL

Uranium-233, James Rushton and Charles Forsberg, ORNL

Heavy Isotopes, Ron Canon and Emory Collins, ORNL

Highly Enriched/Low-Enriched/and Natural Uranium, J. Dale Jackson, ORNL

Pu-238, Robert Wham, ORNL

Tour—DU Uses Research (Heavy Concrete/Catalysis), L. Dole, C. Mattus, S. Dai, ORNL

Tour—Radiochemical Engineering Development Center, B. Patton, J. Knauer, ORNL

Tour—East Tennessee Technology Park DU Storage Cylinders, H. Philpot, ORNL

Augusta, Georgia, March 7-8, 2002

Site Overview and EM Nuclear Materials Planning, Jay Bilyeu, DOE-Savannah River (DOE-SR)

Summary of Site Nuclear Materials Science Needs, Alan Riechman, Westinghouse Savannah River Company (WSRC)

Tour 330-M and 331-M—Depleted Uranium Storage, James Wiederkehr, WSRC, and Dawn Gillas, DOE-SR

Driving tour F Area A-Line—Depleted Uranium Solution Conversion, George Zachmann, WSRC

Tour 717-F Mock-up Shop, Dave Barnette, Bechtel Savannah River Inc.
Tour H-Canyon Control Room, Don Johnson, Glynn Dyer, Jimmy Winkler, WSRC

HB-Line Overview—Nuclear Materials Recovery, Dick Murphy, WSRC

USDOE Nuclear Material Disposal Orphans, Billy Chambers, DOE-SR

DOE-EM Aluminum-Based SNF Disposition Alternatives—Potential for H-Canyon Reprocessing, Billy Chambers, DOE-SR

Radiolysis of Adsorbed Moisture, Neal Askew, WSRC

Gas Generation Research to Support Transportation and Storage of Plutonium-Bearing Materials, Jon Duffy and Ron Livingston, WSRC

Prevention of Precipitation of Unwanted Solids During Nuclear Material Processing, William J. Crooks III, WSRC

Computer Modeling of Uranium and Plutonium Solvent Extraction Processes, Mark Crowder, WSRC

Interaction of Actinide Process Solutions with Concrete, Michael Bronikowski, WSRC

Aqueous Pretreatment of LWR Fuel for Accelerator Transmutation of Waste, Major Thompson, WSRC

Plutonium Storage Science Needs, Kerry Dunn and Thad Adams, WSRC

Test Program of Impact/Crush/Thermally Resistive Materials for Radioactive Materials Packaging, P.S. Blanton and A. C. Smith, WSRC

Remote Monitoring of Nuclear Materials Under Surveillance, Bill Rigot, WSRC

SRS SNF Science Needs, Thad M. Adams, WSRC

Richland, Washington, May 20-22, 2002

Integration of Science at Hanford, John LaFemina and Terry Walton, Pacific Northwest National Laboratory (PNNL)

Plutonium Finishing Plant (PFP) Project Overview, Larry Romine, DOE-Richland Operations Office (DOE-RL)

Spent Nuclear Fuel (SNF) Project Overview, Roger McCormack, Fluor Hanford

Cs/Sr Capsules Overview, George Sanders and Sen Moy, DOE-RL

Waste Management—Central Waste Complex, George Sanders and Todd Shrader, DOE-RL

EM Science Program at Hanford, Paul Bredt, PNNL

INEEL Spent Nuclear Fuels, Phil Winston, INEEL

SNF Technology Gaps, Jim Sloughter and Bruce Makenas, Fluor Hanford, and John Abrehah, PNNL

Non-Destructive Special Nuclear Materials Analysis, Tony Peurrung, PNNL

Nuclear Materials Technology Gaps, Ted Venetz, Fluor Hanford, Cal Delegard and Andy Schmidt, PNNL

PFP Technical Roundtable, Suzanne Clarke, DOE-RL, Lenny Perkins, DFSH, and Dwayne Speer, DOE-RL

Appendix D

List of Acronyms

AES	Auger electron spectroscopy
AFCI	Advanced Fuel Cycle Initiative
ANSTO	Australian National Science and Technology Organization
BNFL	British Nuclear Fuels, Limited
DOE	Department of Energy
DNFSB	Defense Nuclear Facilities Safety Board
DU	depleted uranium
DUF_6	depleted uranium hexafluoride
DWPF	Defense Waste Processing Facility
EM	DOE Office of Environmental Management
EMSP	Environmental Management Science Program
ERSD	DOE Environmental Remediation Sciences Division
HEU	highly enriched uranium
HF	hydrogen fluoride
HLW	high-level waste
IAEA	International Atomic Energy Agency
ICRP	International Commission on Radiological Protection
INEEL	Idaho National Engineering and Environmental Laboratory
LEU	low-enriched uranium
LWR	light water reactor

MFFF	MOX fuel fabrication facility
MOX	mixed oxide fuel
MPC&A	material protection, control and accounting
MTHM	metric tons of heavy metal
NAS	National Academy of Sciences
NDA	nondestructive analysis
NNSA	National Nuclear Security Administration
NRC	National Research Council
NZP	sodium zirconium phosphate
ORNL	Oak Ridge National Laboratory
RFETS	Rocky Flats Environmental Technology Site, Colorado
ROO	DOE Richland Operations Office, Washington
SNF	spent nuclear fuel
SROO	DOE Savannah River Operations Office, South Carolina
SRS	Savannah River Site
SS	stainless steel
TRU	transuranic
USEC	U.S. Enrichment Corporation
USNRC	U.S. Nuclear Regulatory Commission
WAC	waste acceptance criteria
WESF	Waste Encapsulation and Storage Facility
WHO	World Health Organization
WIPP	Waste Isolation Pilot Plant
WSRC	Westinghouse Savannah River Company
WAPS	waste acceptance product specification
XPS	X-ray photoelectron spectroscopy