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Muhammad Sahimi

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From Classical Methods to Modern Approaches

Second, Revised and Enlarged Edition



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Dedicated to the memory of my parents

Habibollah Sahimi (1916–1997) and Fatemeh Fakour Rashid (1928–2006)

and to

Mahnoush, Ali and Niloofar

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Preface to the Second Edition

Since 1995, when the first edition of this book was published, the science of porous media and the understanding of flow and transport phenomena that occur in them have been advanced greatly. Characterization of porous media – both fractured and unfractured – can now be done in great detail, that is, for samples on the laboratory scale. Reconstruction methods have made it possible to develop realistic models of porous media based on relatively limited experimental data. Many new approaches have been developed that have made it possible to compute various flow and transport properties of porous media with considerable precision. In particular, pore network models on the one hand, and advanced computational techniques, such as the lattice-Boltzmann method, on the other hand, have become invaluable tools for studying flow and transport in porous media and in fractures. New theoretical developments have made it possible to analyze field-scale data, such as, the various types of logs and seismic records, with precisions, hence yielding deeper insights.

The understanding of fractures – particularly the crucial effect of the roughness of their internal surface – and fracture networks – especially the effect of their connectivity on their effective (overall) properties – has deepened. Classical models of fractured porous media, for example, the double-porosity model, though still useful in some special limits, are no longer viewed as the only practicable models for simulating flow and transport in large-scale fractured porous media. Viable alternatives that are much more realistic have been emerging at a rapid pace.

On the experimental side, new instrumentations coupled with advanced theoretical developments have made it possible to measure the various morphological, flow, and transport properties of porous media. Use of such techniques as three-dimensional X-ray computed tomography and nuclear magnetic resonance for measuring the properties have almost become routine.

The new developments motivated the preparation of the second edition of the book. However, the new edition does not merely represent an updated version of the first edition. Almost all the chapters have been completely rewritten. The characterization of unfractured and fractured porous media has been separated, each described and developed in its own chapter. The classical continuum models of fluid flow and transport in porous media and the discrete approaches based on the network models have also been separated, with each subject having its own chapter. On the experimental side, many newer experimental techniques for measuring

the important morphological, flow, and transport properties of porous media have been described. Additionally, instead of describing unconsolidated porous media separately, as in the first edition, they have been merged with consolidated porous media and given equal footing in order to describe both types of porous media in a unified manner.

As the famous song by John Lennon and Paul McCartney goes, *I get by with a little help from my friends*, except that in my case, my students and colleagues have given me *a lot* of help. Many people have contributed to my understanding of the topics described in this book. First and foremost, I have been blessed by the many outstanding doctoral students that I have worked with throughout my academic career on the problems studied in this book. They include Drs. Sepehr Arbabi, Mitra Dadvar, Fatemeh Ebrahimi, Jaleh Ghassemzadeh, Hossein Hamzehpour, Mehrdad Hashemi, Abdossalam Imdakm, Mahyar Madadi, Ali Reza Mehraabi, Sumit Mukhopadhyay, Mohammad Reza Rasaei, and Habib Siddiqui. Over the years, I have also been fortunate enough to have fruitful collaborations with many friends and colleagues on research problems related to what is studied in this book, including Professors Joe Goddard, Manouchehr Haghighi, Barry Hughes, Mark Knackstedt, Charles Sammis, Dietrich Stauffer, and Theodore Tsotsis as well as Drs. Adel A. Heiba and Luigi Sartor.

In addition, the preparation of this edition of the book was greatly helped by several people. My former doctoral students, Drs. Faezeh Bagheri-Tar, Fatemeh Ebrahimi, and Mahyar Madadi provided much needed help for the figures. Dr. Mohammad Piri generously gave me the electronic file of his outstanding Ph.D. Thesis (Piri, 2003) that I used as a great source for the discussions of multiphase flows as well as the electronic files of the figures, some of which I have used in Chapter 15.

Throughout my academic life, I have been blessed by great mentors. Dr. Hasan Dabiri, my first academic mentor when I was attending the University of Tehran in Iran, introduced me to the petroleum industry when he worked with me on the project for my B.S. degree, *Evaporation Loss in the Petroleum Industry*. Over 35 years after taking the first of many courses with him, I am still influenced by his outstanding qualities, both as an academic mentor and as a wonderful human being. My advisors for my Ph.D. degree at the University of Minnesota, the late Professors H. Ted Davis and L.E. (Skip) Scriven, introduced me to various porous media problems and percolation theory, and taught me the fundamental concepts.

Michael C. Poulson was the publisher of the first edition of this book (and my first book, *Applications of Percolation Theory*, when he was with Taylor and Francis) as well as a great friend. He passed away on December 31, 1996 at the age of 50. In preparing this edition of the book, I greatly missed his wise advice, humor, and cheerful personality. The staff of Wiley-VCH, particularly Ulrike Werner, were extremely patient with my long delay in delivering the 2nd edition.

I dedicate this book to the special people in my life. My mentors in life, my late parents, Habibollah Sahimi and Fatemeh Fakour Rashid, made me what I am. I will miss them until I see them again. My wife Mahnoush, son Ali, and daughter Nilofar put up with my long absence from family life, and my spending countless

numbers of days, weeks, and months in front of the computer at home. This edition would not have been completed without their love and patience.

Los Angeles, August 2010

Muhammad Sahimi

Preface to the First Edition

Disordered porous media are encountered in many different branches of science and technology, ranging from agricultural, ceramic, chemical, civil and petroleum engineering, to food and soil sciences, and powder technology. For several decades, porous media have been studied both experimentally and theoretically. With the advent of precise instruments and new experimental techniques, it has become possible to measure a wide variety of physical properties of porous media and flow, and transport processes therein. New computational methods and technologies have also allowed us to model and simulate various phenomena in porous media, and thus a deep understanding of them has been gained.

Whether we like it or not, we have to accept the fact that many natural porous systems are fractured, the understanding of which requires new methodologies and ways of thinking. In the past two decades, the understanding of fractured rock has taken on new urgency since, in addition to oil reservoirs, many groundwater resources are also fractured. Thus, flow in fractured rock has attracted the attention of scientists, engineers and *politicians* as a result of growing concerns regarding pollution and water quality. Highly intense exploitation of groundwater, and the increase in solute concentrations in aquifers due to leaking repositories and use of fertilizers, have made flow in fractured rock a main topic of research.

I have been working on such problems for 15 years, and during this period, I have realized that there are two distinct approaches to modeling flow phenomena in porous media and fractured rock. Some of these approaches belong to a class of models that I call the *continuum models*. Largely based on the classical equations of flow and transport, the continuum models have been very popular with engineers. Although not as widely used as the continuum models, the second approach, which is based on the *discrete models* that represent a porous system by a discrete set of elements and use large-scale Monte Carlo simulations and various statistical methods to analyze flow phenomena in porous media and fractured rock, has also attracted wide attention. Many new ideas and concepts have been developed as the result of using this class of models, and new results have emerged that have helped us gain a much better understanding of porous systems. In addition, such ideas and concepts as percolation processes, universal scaling laws and fractals, the basic tools of the discrete models, have gradually found their rightful positions in the porous systems literature. Currently, such concepts are even taught in graduate

courses on flow through porous media, and in courses on computer simulation of disordered media and statistical mechanical systems.

Realizing these facts, and given that there was no book that discusses and compares *both* approaches, I decided to write this book. Even a glance at the immense literature on these subjects reveals that it is impossible to discuss every issue and present an in-depth analysis of it in just one book. Percolation theory, fractals, Monte Carlo simulations, and similar topics have been, by themselves, the subjects of several books and monographs. Unless one explains the most important concepts and then provides references where the reader can find more material to read, such a book can easily contain over a thousand pages. Based on this realization and limitation, I selected the topics that are discussed in this book. Largely based on this limitation, I also had to ignore several important topics, for example, flow of non-Newtonian fluids in porous media, filtration, and dissolution of rock by an acid which creates large fractures in the rock. In spite of such limitations, this book represents, in my opinion, a comprehensive review and discussion of the most important experimental and theoretical approaches to flow phenomena in porous media and fractured rock. Therefore, it can be used both as a reference book and a text for graduate courses on the subjects that it discusses. Considering the fact that this book discusses experimental measurement of the most important morphological and transport properties of porous systems, and the fact that many topics, especially those of single-phase flow and transport, are discussed in great detail, I believe that roughly half of the book can also be used in a senior-level undergraduate course on porous media problems that is taught in many chemical, petroleum and civil engineering and geological science departments.

As the famous song by John Lennon and Paul McCartney goes, "I get by with a little help from my friends", except that in my case, my friends and colleagues have given me *a lot* of help. Many people have contributed to my understanding of the topics discussed in this book, a complete list of whom would be too long to be given here. However, I would like to mention a few of them who have had greatly influenced my way of thinking. I would like to thank Professors H. Ted Davis and L. E. (Skip) Scriven of the University of Minnesota who introduced me to various porous media problems and percolation theory, and taught me the fundamental concepts when I was their doctoral student. For over a decade, Dietrich Stauffer has greatly influenced my way of thinking about percolation, disordered media, and critical phenomena. I am deeply grateful to him. I would like to thank all of my past collaborators with whom I have published many papers on flow in porous media, especially Adel A. Heiba and Barry D. Hughes. Three other persons helped me write and finish this book. Michael Poulson, my publisher at VCH Publishers, was very patient and helpful. Drs. Sherry Caine and Dalia Goldschmidt helped me to organize my thoughts, focus on writing this book, and have a more positive outlook on life.

My debts of gratitude to them, and to many more who taught and influenced me, thus making this book possible.

Los Angeles, August 1994

Muhammad Sahimi

1

Continuum versus Discrete Models

Introduction

Flow and transport phenomena in porous media and fractured rock as well as industrial synthetic porous matrices arise in many diverse fields of science and technology, ranging from agricultural, biomedical, construction, ceramic, chemical, and petroleum engineering, to food and soil sciences, and powder technology. Fifty percent or more of the original oil-in-place is left behind in a typical oil reservoir after the primary and secondary recovery processes end. The unrecovered oil is the main target for the enhanced or tertiary oil recovery methods now being developed. However, oil recovery processes only constitute a small fraction of an enormous and still rapidly growing literature on porous media. In addition to oil recovery processes, the closely related areas of soil science and hydrology are perhaps the best-established topics related to porous media. The study of groundwater flow and the restoration of aquifers that have been contaminated by various pollutants are important current areas of research. The classical research areas of chemical engineers that deal with porous media include filtration, centrifuging, drying, multiphase flow in packed columns, flow and transport in microporous membranes, adsorption and separation, and diffusion and reaction in porous catalysts.

Lesser known, though equally important, phenomena involving porous media are also numerous. For example, for the construction industry, the transmission of water by building materials (bricks or concretes) is an important problem to consider when designing a new building. The same is true for road construction where penetration of water into asphaltene damages roads. Various properties of wood, an interesting and unusual porous medium, have been studied for a long time. Some of the phenomena involving wood include drying and impregnation by preservatives. Civil engineers have long studied asphalts as water-resistant binders for aggregates, protection of various types of porous materials from frost heave, and the properties of road beds and dams with respect to water retention. Biological porous media with interesting pore space morphology and wetting behavior include skin, hair, feathers, teeth and lungs. Other types of porous media that are widely used are ceramics, pharmaceuticals, contact lenses, explosives, and printing papers.

In any phenomenon that involves a porous material, one must deal with the complex pore structure of the medium and how it affects the distribution, flow, displacement of one or more fluids, or dispersion (i.e., mixing) of one fluid in another. Each process is, by itself, complex. For example, displacement of one fluid by another can be carried out by many different mechanisms which may involve heat and mass transfer, thermodynamic phase change, and the interplay of various forces such as viscous, buoyancy, and capillary forces. If the solid matrix of a porous medium is deformable, its porous structure may change during flow or some other transport phenomenon. If the fluid is reactive or carries solid particles of various shapes, sizes, and electrical charges, the pore structure of the medium may change due to the reaction of the fluid with the pore surface, or the physicochemical interaction between the particles and the pore surface.

In this book, we describe and study various experimental, theoretical, and computer simulation approaches regarding diffusion, flow, dispersion, and displacement processes in porous media and fractured rock. Most of the discussions regarding porous media are equally applicable to a wide variety of systems, ranging from oil reservoirs to catalysts, woods, and composite materials. We study flow phenomena only in a *static* medium, that is, one with a morphology that does not change during a given process. Thus, deformable media as well as those that undergo morphological changes due to a chemical reaction, or due to physicochemical interactions between the pore surface and a fluid and its contents, are not studied here. The interested reader is referred to Sahimi *et al.* (1990) for a comprehensive discussion of transport and reaction in evolving porous media and the resulting changes in their morphology.

1.1

A Hierarchy of Heterogeneities and Length Scales

The outcome of any given phenomenon in a porous medium depends on several length scales over which the medium may or may not be homogeneous. By homogeneous, we mean a porous medium with effective properties that are *independent* of its linear size. When there are inhomogeneities in the medium that persist at distinct length scales, the overall behavior of the porous medium is dependent on the rate of the transport processes, for example, diffusion, conduction, and convection, the way the fluids distribute themselves in the medium, and the medium's morphology. Often, the morphology of a porous medium plays a role that is more important than that of other influencing factors.

Consider, as an example, an oil reservoir, perhaps one of the the most important heterogeneous porous media. In principle, the reservoir is completely deterministic in that it has potentially measurable properties and features at various length scales. It could have been straightforward to obtain a rather complete description of the reservoir if only we could excavate each and every part of it. In practice, however, this is not possible and, therefore, a description of any reservoir, or any other natural porous medium for that matter, is a combination of the determinis-

tic components – the information that can be measured – and indirect inferences that, by necessity, have stochastic or random elements in them. Over the past four decades, the statistical physics of disordered media has played a fundamental role in developing the stochastic component of description of porous media.

There are several reasons for the development. One is that the information and data regarding the structure and various properties of many porous media are still vastly incomplete. Another reason is that any property that we ascribe to a medium represents an average over some *suitably selected volume* of the medium. However, the relationship between the property values and the volume of the system over which the averages are taken remains unknown. The issue of a suitably selected volume reminds us that any proper description of a porous medium or fractured rock must have a length scale associated with it. In general, the heterogeneities of a natural porous medium are described at mainly four distinct length scales that are as follows (Haldorsen and Lake, 1984).

1. The *microscopic heterogeneities* are at the level of the pores or grains, and are discernible only through scanning electron microscopy or thin sections.
2. The *macroscopic heterogeneities* are at the level of core plugs, and are routinely collected in fields and analyzed. Such heterogeneities are found in every well with property values varying widely from core to core. In most theoretical studies, however, cores are assumed homogeneous and the average effective properties are assigned to them, notwithstanding their microscopic heterogeneities.
3. The *megascopic* or *field-scale heterogeneities* are at the level of the entire reservoir that may have large fractures and faults. They can be modeled as a collection of thousands, perhaps millions, of cores, oriented and organized in some fashion.
4. The *gigascopic heterogeneities* are encountered in landscapes that may contain many such reservoirs as described in the third example, along with mountains, rivers, and so on.

Not all of the aforementioned heterogeneities are important to all porous media. For example, porous catalysts usually only contain microscopic heterogeneities, and packed beds may be heterogeneous at both the microscopic and macroscopic levels. In this book, we consider the first three classes of heterogeneities and their associated length scales.

1.2

Long-Range Correlations and Connectivity

In the early years of studying flow phenomena in porous media and fractured rock, most researchers almost invariably assumed that the heterogeneities in one region or segment of the system were random and uncorrelated with those in other regions. Moreover, it was routinely assumed that such heterogeneities occur at length scales much smaller than the overall linear size of the system. Such assumptions were partly due to the fact that it was very difficult to model the system in a more

realistic way due to the computational limitations and lack of precise experimental techniques for collecting the required information. At the same time, the simple conceptual models, such as random heterogeneities, did help us gain a better understanding of some of the issues. However, increasing evidence suggests that rock and soils do not conform to such simplistic assumptions. They exhibit *correlations* in their properties, and such correlations are often present at *all* the length scales. The existence of such correlations has necessitated the introduction of *fractal distributions* that tell us how property values of various regions of a porous medium depend on the length (or even time) scale of the observations, how they are correlated with one another, and how one can model such correlations realistically. Such concepts and modeling techniques are described in this book.

Once we accept that natural porous media and fractured rock are heterogeneous at many length scales, we also have to live with its consequences. As a simple, yet very important, example, consider the permeability of a porous medium, which is a measure of how easily a fluid can flow through it. In a natural porous medium and at large length scales (of the order of a few hundred meters or more), the permeabilities of various regions of the medium follow a broad distribution. That is, while parts of the medium may be highly permeable, other parts can be practically impermeable. If we consider a natural porous medium, then, the low permeability regions can be construed as the impermeable zones as they contribute little or nothing to the overall permeability, while the permeable zones provide the paths through which a fluid flows. Thus, the impermeable zones divide the porous medium into compartments according to their permeabilities. This implies that the permeable regions may or may not be connected to one another, and that there is disorder in the connectivity of various regions of the porous medium. Thus, if we are to develop a realistic description of a porous medium, the connectivity of its permeable regions must be taken into account.

The language and the tools for taking into account the effect of the connectivity of the permeable regions of a pore space are provided by *percolation theory*. Similar to fractal distributions, percolation has its roots in the mathematics and physics literature, although it was first used by chemists for describing polymerization and gelation phenomena. Percolation theory teaches us how the connectivity of the permeable regions of a porous medium affects its overall properties. Most importantly, percolation theory predicts that if the volume fraction of the permeable regions is below some critical value, the pore space is not permeable and its overall permeability is zero.

In the classical percolation that was studied over 50 years ago, it was assumed that the permeable and impermeable regions are distributed randomly and independently of each other throughout the pore space. Since then, more refined and realistic percolation models have been developed for taking into account the effect of correlations and many other influencing factors. Such ideas and concepts are developed and used throughout this book for describing various flow phenomena in porous media and fractured rock.

1.3

Continuum versus Discrete Models

Now that we know what kinds of heterogeneities one must deal with in studying porous media, it is also necessary to consider the types of models that have been developed over the past several decades for describing flow and transport phenomena in porous media and fractured rock. The analysis of flow, dispersion, and displacement processes in porous and fractured media has a long history in connection with the production of oil from underground reservoirs. It was, however, only in the past 35 years that the analysis has been extended to include detailed structural properties of the media. Such studies are quite diverse in the physical phenomenon that they consider. In this book, we divide the models for flow, dispersion, and displacement processes in porous and fractured media into two groups: the *continuum models* and *discrete* or *network models*.

Continuum models represent the classical engineering approach to describing materials of complex and irregular geometry characterized by several distinct relevant length scales. The physical laws that govern flow and transport at the microscopic level are well understood. Thus, one can, in principle, write down differential equations for the conservation of momentum, energy, and mass and the associated initial and boundary conditions at the fluid-solid interface. However, as the interface in typical porous media is very irregular, practical and computationally and economically feasible techniques, while available, are often not feasible for solving such boundary-value problems – even when one knows the detailed morphology of the porous medium. Determination of the precise solid-fluid boundary remains a very difficult (if not impossible) task, particularly for large-scale porous media. The boundary within which one would have to solve the equations of change are so tortuous as to render the problem mathematically intractable. Moreover, even if the solution of the problem could be obtained in great detail, it would contain much more information than would be useful in any practical sense. Thus, it becomes essential to adopt a macroscopic description at a length scale much larger than the dimension of individual pores or fractures.

Effective properties of a porous medium are defined as averages of the corresponding microscopic values. The averages must be taken over a volume that is small enough compared with the volume of the system, but large enough for the equation of change to hold when applied to that volume. At every point in the medium, one uses the smallest such volume, thereby generating macroscopic field variables satisfying such equations as Darcy's law of flow or Fick's law of diffusion. The reasons for choosing the smallest suitable volume for averaging are to allow in the theory suprapore variations of the porous medium and to generate a theory capable of treating the usual macroscopic variations of the effective properties. In this book, we encounter several situations where the conditions for the validity of such an averaging are not satisfied. Even when the averaging is theoretically sound, the prediction of the effective properties is often difficult because of the complex structure of the pore space. In any case, with empirical, approximate, or exact formulae for the flow and transport coefficients and other effective

properties, the consequences of a given phenomenon in a porous medium can be analyzed based on such a theory. As mentioned above, many of the past theoretical attempts to derive effective flow and transport coefficients of porous media from their microstructure entailed a simplified representation of the pore space, often as a bundle of capillary tubes. In this model, the capillaries were initially treated as parallel, and then later as randomly oriented. Such models are relatively simple, easy to use, and sufficiently accurate, provided that the relevant parameters are determined experimentally and the connectivity of the pore space does not play a major role. Having derived the effective governing equations and suitable flow and transport properties, one has the classical description of a porous medium as a continuum. We shall, therefore, refer to various models associated with the classical description as the continuum models.

The continuum models have been widely used due to their convenience and familiarity to the engineer. They do have some limitations, one of which was noted earlier in the discussion concerning scales and averaging. They are also not well-suited for describing those phenomena in which the connectivity of the pore space or the fracture network, or that of a fluid phase, plays a major role. Continuum models also break down if there are correlations in the system with an extent that is comparable with the linear size of the porous medium.

The second class of models, the *discrete models*, are free of the limitations of the continuum models. They have been advanced to describe phenomena at the microscopic level and have been extended in the last decade or so years to describe various phenomena at the macroscopic and even larger scales. Their main shortcoming, from a practical point of view, is the large computational effort required for a realistic discrete treatment of the pore space. They are particularly useful when the effect of the pore space or fracture network connectivity, or the long-range correlations, is strong. The discrete models that we consider in this book are mostly based on a network representation of porous media and fracture networks. The original idea for network representation of a pore space is rather old and goes back to the early 1950s, but it was only in the early 1980s that systematic and rigorous procedures were developed to map, in principle, any disordered porous medium onto an equivalent network. Once the mapping is complete, one can study a given phenomenon in porous media in great detail.

However, only in the past 35 years have ideas from the statistical physics of disordered media been applied to flow, dispersion, and displacement processes in porous and fractured media. The concepts include percolation theory, and fractal distributions and structures that are the main tools for describing the scale-dependence of the effective properties of disordered media and how long-range correlations affect them. What we intend to do in this book is to describe and review the relevant literature on the subject, define and discuss the ideas and techniques from the statistical physics of disordered media and their applications to the processes of interest in this book, and describe the progress that has been made as a result of such applications. In particular, we emphasize the important effect of the connectivity of the pores or fractures of a porous medium on the phenomena

of interest. We also describe the characterization of fractured porous media and flow and transport in the fracture networks in great detail.

In summary, within this book we study models of porous media and fractured rock, explain various experimental techniques that are used for characterizing their morphology and flow and transport therein, describe the continuum models of flow and transport in such pore media, and compare them with the predictions of the discrete models. In all cases, we contrast the classical models and techniques with the modern approaches based on the discrete models. As such, we believe that the book is unique, as it treats the subjects of porous media and fractured rock on equal footing. We hope that this book can give the reader a clear view of where we stand in the middle of 2010.

2

The Equations of Change

Introduction

The purpose of this chapter is to present the equations of change – the mass and momentum conservation equations as well as the continuity equation for individual chemical species. The equations presented in this chapter represent the starting point for the analysis of flow and transport processes studied in the rest of this book and, therefore, it may be useful to summarize them here. We do not derive the equations, as the derivations are found in any standard book on transport phenomena, but only summarize the differential forms of the equations of change, as their macroscopic or integral forms are not used in this book. We assume that the reader is familiar with basic vector and tensor calculus. We follow the classical book of Bird *et al.* (2007), first published in 1960, that the author has used over the past four decades, both as a student and as a teacher. Other readable accounts of the equations are given by Aris (1962) and Batchelor (1967).

2.1

The Mass Conservation Equation

Suppose that ρ is the density of a flowing fluid and $\mathbf{v}$ is its velocity vector at time t . Then, the differential equation that is the mathematical statement for the physical fact that the total mass is conserved is given by

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{v}) = 0, \quad (2.1)$$

where ∇ is the “del” operator, and $\nabla \cdot (\rho \mathbf{v})$ is the *divergence* of $\rho \mathbf{v}$. For example, in Cartesian coordinates,

$$\nabla = \mathbf{e}_x \frac{\partial}{\partial x} + \mathbf{e}_y \frac{\partial}{\partial y} + \mathbf{e}_z \frac{\partial}{\partial z}, \quad (2.2)$$

where $\mathbf{e}_x$, $\mathbf{e}_y$, and $\mathbf{e}_z$ are unit vectors in the corresponding directions. In cylindrical coordinates, one has

$$\nabla = \mathbf{e}_r \frac{\partial}{\partial r} + \mathbf{e}_\theta \frac{1}{r} \frac{\partial}{\partial \theta} + \mathbf{e}_z \frac{\partial}{\partial z}, \quad (2.3)$$

where $\mathbf{e}_r$, $\mathbf{e}_\theta$, and $\mathbf{e}_z$ are the corresponding unit vectors, while in spherical coordinates the del operator is given by

$$\nabla = \mathbf{e}_r \frac{\partial}{\partial r} + \mathbf{e}_\theta \frac{1}{r} \frac{\partial}{\partial \theta} + \mathbf{e}_\phi \frac{1}{r \sin \theta} \frac{\partial}{\partial \Phi} . \quad (2.4)$$

If we use the *substantial derivative* operator

$$\frac{D}{Dt} = \frac{\partial}{\partial t} + \mathbf{v} \cdot \nabla , \quad (2.5)$$

then, the equation of continuity can be rewritten as

$$\frac{D\rho}{Dt} + \rho \nabla \cdot \mathbf{v} = 0 . \quad (2.6)$$

For an incompressible fluid, ρ is constant, and Eq. (2.6) reduces to

$$\nabla \cdot \mathbf{v} = 0 . \quad (2.7)$$

2.2

The Momentum Equation

Suppose that P is the pressure, $\boldsymbol{\tau}$ the stress tensor, and $\mathbf{g}$ the gravitational acceleration vector. Then, the momentum equation is given by

$$\frac{\partial(\rho \mathbf{v})}{\partial t} = -\nabla \cdot (\rho \mathbf{v} \mathbf{v}) - \nabla \cdot \boldsymbol{\tau} - \nabla P + \rho \mathbf{g} . \quad (2.8)$$

Observe that the first term on the right side of Eq. (2.8) is the rate of convective momentum, whereas the second term represents the rate of viscous momentum, both per unit volume of the system. The term $\mathbf{v} \mathbf{v}$ is a *dyadic product* of the two vectors that, similar to $\boldsymbol{\tau}$, is a second-rank tensor. $\nabla \cdot \boldsymbol{\tau}$ and $\nabla \cdot (\rho \mathbf{v} \mathbf{v})$ are, therefore, not simple divergences. Moreover, the stress tensor is symmetric. What distinguishes a Newtonian fluid from a non-Newtonian one is the form of the stress tensor $\boldsymbol{\tau}$. For a Newtonian fluid and in Cartesian coordinates, the components of the stress tensor are given by

$$\tau_{\zeta\zeta} = -2\mu \frac{\partial v_\zeta}{\partial \zeta} + \frac{2}{3}\mu (\nabla \cdot \mathbf{v}) , \quad \zeta = x, y, z , \quad (2.9)$$

$$\tau_{\zeta\xi} = -\mu \left(\frac{\partial v_\zeta}{\partial \xi} + \frac{\partial v_\xi}{\partial \zeta} \right) , \quad (\zeta, \xi) = (x, y), (y, z), (z, x) , \quad (2.10)$$

where μ is the viscosity of the fluid. Similar expressions for the components of $\boldsymbol{\tau}$ in cylindrical and spherical coordinates are given by Bird *et al.* (2007). In terms of the substantial derivative, the equation of motion is written as

$$\rho \frac{D\mathbf{v}}{Dt} = -\nabla \cdot \boldsymbol{\tau} - \nabla P + \rho \mathbf{g} . \quad (2.11)$$

If the density and viscosity of the fluid are constant, then

$$\rho \frac{D\mathbf{v}}{Dt} = \mu \nabla^2 \mathbf{v} - \nabla P + \rho \mathbf{g} , \quad (2.12)$$

which is the well-known Navier¹⁾–Stokes²⁾ equation. If the viscous effects are negligible, then we obtain the Euler³⁾ equation:

$$\rho \frac{D\mathbf{v}}{Dt} = -\nabla P + \rho \mathbf{g} , \quad (2.13)$$

which is used for describing flow of an inviscid fluid. If the inertial effects, represented by $\rho D\mathbf{v}/Dt$, are negligible, which is often the case for the flow phenomena studied in this book (for which the Reynolds number is very small), we obtain the Stokes' equation,

$$-\nabla P + \mu \nabla^2 \mathbf{v} + \rho \mathbf{g} = \mathbf{0} , \quad (2.14)$$

which is used heavily throughout this book.

2.3

The Diffusion and Convective-Diffusion Equations

We consider a binary mixture of two miscible fluids, one of which is the solvent, while the other one is the solute. Suppose that the concentration of the solute is C , and that the molecular diffusivity of the solute in the solvent is D_m . Then, the continuity equation for the solute is given by

$$\frac{\partial C}{\partial t} + \nabla \cdot \mathbf{J} = R_A , \quad (2.15)$$

where R_A is the molar rate of reaction (if there is any) per unit volume, and $\mathbf{J}$ the total flux of the solute, given by

$$\mathbf{J} = C\mathbf{v} - D_m \nabla C . \quad (2.16)$$

Therefore, the equation of continuity for the solute is given by

$$\frac{\partial C}{\partial t} + \nabla \cdot (C\mathbf{v}) = \nabla \cdot (D_m \nabla C) + R_A . \quad (2.17)$$

1) *Claude Louis Marie Henri Navier* (1785–1836) was a French civil engineer and physicist whose specialty was mechanics, and in particular, road and bridge building. He formulated the general theory of elasticity. He is considered to be the founder of modern structural analysis.

2) *George Gabriel Stokes* (1819–1903) was a mathematician and physicist who taught at Cambridge University and was president of the Royal Society. He made important

contributions to fluid dynamics, optics, and mathematical physics. In addition to Navier–Stokes equations, Stokes' law, Stokes' theorem, Stokes' line, Stokes' relations, and Stokes' shift are all named in his honor.

3) *Leonhard Euler* (1707–1783) (pronounced "Oiler") was a Swiss born mathematician who taught in St. Petersburg, Basel, and Berlin, and published many papers in many areas of mathematics and physics.

A similar equation can be written for the solvent. If both $\mathbf{v}$ and D_m are constant, and if the fluids are not reactive, then

$$\frac{\partial C}{\partial t} + \mathbf{v} \cdot \nabla C = D_m \nabla^2 C, \quad (2.18)$$

which is the well-known *convective-diffusion equation*, heavily used in Chapters 11–13. Here, ∇^2 is the Laplacian operator that, in Cartesian coordinates, is given by

$$\nabla^2 = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2}, \quad (2.19)$$

while in cylindrical coordinates we have

$$\nabla^2 = \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial}{\partial r} \right) + \frac{1}{r^2} \frac{\partial^2}{\partial \theta^2} + \frac{\partial^2}{\partial z^2}, \quad (2.20)$$

and in spherical coordinates one has

$$\nabla^2 = \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial}{\partial r} \right) + \frac{1}{r^2 \sin \theta} \frac{\partial}{\partial \theta} \left(\sin \theta \frac{\partial}{\partial \theta} \right) + \frac{1}{r^2 \sin^2 \theta} \frac{\partial^2}{\partial \Phi^2}. \quad (2.21)$$

If $R_A = 0$ and the fluids are stagnant, then we obtain the well-known diffusion equation

$$\frac{\partial C}{\partial t} = \nabla \cdot (D_m \nabla C). \quad (2.22)$$

More generally, instead of a single-valued diffusivity, one may have an *effective diffusivity tensor*. For example, if a porous medium is anisotropic, then each principal direction of the system is characterized by a distinct diffusivity. Even if the medium is not structurally anisotropic, the overall behavior of the transport of the solute in the solvent may be characterized by an effective diffusivity tensor. Dispersion phenomena that are studied in Chapters 11 and 12, and are also important to miscible displacements that are studied in Chapter 13, provide an example of a system in which there is a *flow-induced* dynamical anisotropy and, thus, one needs more than one effective diffusivity to characterize the phenomena.

2.4

Fluid Flow in Porous Media

The conservation equations must be supplemented by additional correlations by which one can calculate physical properties of the fluids, for example, their viscosities, densities and diffusivities. Moreover, for flow through porous media, one needs to relate macroscopic and measurable quantities such as the average fluid velocity, to the morphological properties of the media. For flow through porous

media, the Darcy's law relates the fluid velocity to the permeability K of a porous medium

$$\mathbf{V} = -\frac{K}{\mu}(\nabla P - \rho \mathbf{g}) . \quad (2.23)$$

The permeability K depends on the porosity ϕ of the medium, that is, the volume fraction of its pore space, and a major task is to predict K for a given porous medium. These matters will be studied in Chapters 9–12. Moreover, when the equations are used for a porous medium, one must take into account the fact that only a fraction ϕ of the medium is actually used for flow, diffusion and dispersion. Thus, for example, Eq. (2.17) should be rewritten as

$$\frac{\partial(\phi C)}{\partial t} + \nabla \cdot (\phi C \mathbf{v}) = \nabla \cdot (\phi D_m \nabla C) + R_A . \quad (2.24)$$

In most cases studied in this book, the boundary conditions are either of the *Dirichlet type* in which the value of the unknown on the boundaries or a portion of them is specified, or are of the *Neumann type* in which the flux of the quantity in the direction normal to the external surface of the system is specified. The effect of the length scale for macroscopic homogeneity of a given porous medium and the correlations between various regions of the system are also important. How they affect the mathematical form of the continuum equation of motion for describing a flow phenomenon in a porous medium is an important subject in this book.

3

Characterization of Pore Space Connectivity: Percolation Theory

Introduction

For many decades, a porous medium was imagined and modeled as a bundle of tubes, either in parallel or series, presumably due to the misleading similarity between the Darcy's law that described slow flow of fluid in a porous medium and the Hagen–Poiseuille law for laminar flow through capillary tubes. Even if one considers the flow of a Newtonian fluid, the analogy between the two laws is not as sound as it may seem, as the linearity of the Darcy's law is due to the *assumption* of flow being in the creeping regime, whereas the Poiseuille flow is *unconditionally* linear (Philip, 1970). Despite the obvious shortcomings of representing porous media by a bundle of tubes, many formulae were derived for the effective permeability based on such models. However, such formulae failed to capture the essential physics of flow and transport through porous media, and to provide quantitative predictions for most, if not all, of the quantities of interest. It was, therefore, recognized that the reason for the utter failure of the bundle of pores model is its inability for accounting for the fact that the pores in most porous media are not simply arranged in parallel series, rather they are distributed chaotically in the space, and are *interconnected*. Hence, researchers' attention turned towards developing realistic models of porous media that can account for the effect of the interconnectivity of the pores. The first systematic attempt in this direction was perhaps made by Fatt (1956a,b,c)¹⁾, who carried out the research as part of his Ph.D. dissertation at the University of Southern California, where the author works.

3.1

Network Model of a Porous Medium

Fatt recognized that, in reality, most porous media – especially natural ones – represent interpenetrating networks of void space and solid matrix. The morphology

1) *Irving Fatt* (1920–1996) was professor of engineering science in the College of Engineering and of physiological optics in the School of Optometry at the University of California in Berkeley. He made fundamental contributions to flow in porous media, flow of oxygen and CO₂ through the cornea, and tears around contact lenses.

of such porous media – the shapes and sizes of the voids, the way they are connected, and the structure of their surface – is usually highly chaotic. The cross section of the voids varies, and the number of ways out of a void – what we will refer to throughout this book as the *coordination number* or *connectivity* – differs from void to void. The internal surface of the voids is usually very rough, highly nonuniform in curvature, and sometimes even varies greatly in chemical composition. The exact description of the entire system is very difficult and (needlessly) elaborate, even now that modern computers are very fast and can handle gigabytes of data. Even if an exact description of a porous medium is possible, one does not need all the details that such a description provides. Indeed, the minimum description that one needs of a porous medium for modeling the essential physics of a given phenomenon depends on the process itself.

For example, to model entrapment of the oil during the primary production of an oil reservoir and its displacement by injecting water and/or gas into the reservoir, the most important feature of the pore space is the connectivity of its pores – or its *topology* – which can be described by an *equivalent network* of *pore throats* – the narrow passages through which the fluids flow – and *pore bodies* – the large voids where the throats meet. For brevity, hereafter we refer to pore bodies and pore throats as pores and throats, respectively. In addition, the relationship between the pores' and throats' sizes, which is part of the *geometry* of the porous medium, is important. In such a network representation of a porous medium, the throats are represented by the *bonds* that are connected to one another at the *sites* or *nodes* of the network that represent the pores. Given a porous medium, it is, in principle, possible to systematically develop its equivalent network and the distribution of the characteristic radii, although there is a degree of arbitrariness in deciding where elementary branch points or nodes (sites) are close enough together to count as multiple branchings, and which pore bodies are so small as not to warrant counting (Mohanty, 1981; Lin and Cohen, 1982). We follow Mohanty (1981) and briefly describe how the network equivalent of a given porous medium is developed.

The idea is to imagine growing, at each point of the wall, a tangent sphere until it makes contact with another wall point, and then joining all of the centers of such inscribed spheres. At points on the edges and at corner points, all spheres must be considered that would be tangent, were the edge or corner infinitesimally rounded. The loci of the centers of two-point-of-contact-spheres define continuous strips that are edged and ended by curves defined by the centers of three-point-of-contact-spheres. Such strips run throughout in the void space and share its connectivity and, therefore, its topology. Moreover, a stripwork (that is, a network of strips) can be reduced to a set of curves by shrinking it along the geodesic normal to its boundaries. The curves are made to carry the information about the local extrema of inscribed sphere radius on the corresponding stripwork, and they, of course, share its topology. Where strips-ends join, so do their curves defining the branch points of the total curves. The total curve defines a virtually unique network equivalent of a porous medium, and is called the *primitive network*.

Branch points on the total curve between which there is no sufficiently deep local minimum of inscribed-sphere radius are clumped together into the nodes or

sites. The criterion to do so is necessarily somewhat arbitrary, and depends on the consideration that are given to the microporosity and roughness of the voids' walls. Then, the throats are the set of points on the curve at which there is a sufficiently deep local minimum of inscribed-sphere radius, as compared with distance between branch points (other ways of defining a throat are also possible). Therefore, there is a branch between every pair of connected nodes. In terms of the terminology that is used to describe actual networks, there is a bond between every pair of directly communicating sites. Each bond is associated with a specific throat and the *locally minimum* radius r_t of its inscribed sphere. Likewise, each site is associated with a specific pore and the *largest* of the *locally maximum* radii r_b of the inscribed sphere. This curve defines the *working network* equivalent of a porous medium. Figure 3.1 shows part of the stripwork and the working network of a porous medium. The innate irregularity of both the void space and the solid matrix of a porous medium has two consequences:

1. The equations of flow and transport can almost never be solved analytically for such porous media. Closed form solutions have only been obtained for porous materials with aligned spherical or elliptical voids (see Chapter 9 and Sahimi, 2003a). One must resort to approximations or computer simulation. These will be described throughout this book.
2. The morphology becomes amenable to statistical analysis. Hence, if the detailed network map of values of r_b and r_t can be obtained, it is used. Otherwise, the map is replaced by the statistical distributions of these quantities. In reality, the local coordination number Z is also a statistically-distributed quantity, but its distribution $f(Z)$ is sometimes approximated by a delta function centered at the *mean* coordination number $\langle Z \rangle$, $f(Z) = \delta(Z - \langle Z \rangle)$. How all such properties are estimated based on experimental data is the subject of the next few chapters.

It is clear that if the throats and pores are not connected enough to form at least one connected path between two opposite external surfaces of a porous medium, no fluid can flow or no transport process can occur. Therefore, one needs a tool, or a set of tools, in order to quantify the effect of pores' interconnectivity on the effective properties of a porous medium. The description of such tools is the subject of the rest of the present chapter.

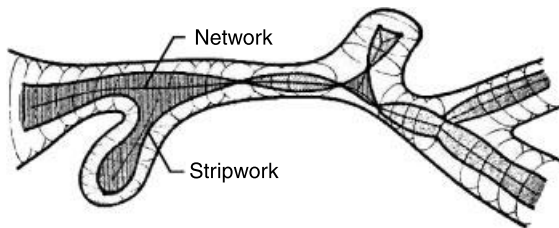


Figure 3.1 A portion of a pore space mapped onto its stripwork and the working network (after Mohanty, 1981).

3.2

Percolation Theory

An important tool for quantifying the effect of the interconnectivity of a pore space on its flow and transport properties is percolation theory. In addition, as we see later in this book, the concepts of percolation theory are essential to the correct interpretation of experimental data obtained by the traditional methods of characterizing porous materials, such as, mercury porosimetry and sorption isotherms.

Percolation theory was first implicitly utilized by Flory (1941)²⁾ and Stockmayer (1943)³⁾ to describe polymerization, a phenomenon in which monomers and small branching molecules react and form large macromolecules. Flory and Stockmayer did not use the word percolation to describe their theory, but the effect of the connectivity of the monomers and small collections of them on the macroscopic properties of a polymer gel was the major new idea in their theory, which is also the major concept in percolation theory. In addition, to derive analytical formulae for various properties of a gel, Flory and Stockmayer ignored the formation of closed loops of reacted monomers, and only considered branching structures. Today, we recognize their work to be equivalent to the description of a percolation process on a special kind of network, namely, the Bethe lattice, an endlessly-branching structure without any closed loops, an example of which is shown in Figure 3.2.

The “modern” percolation theory was introduced by Broadbent and Hammersley (1957)⁴⁾. They were dealing with the problem of the spread of a “fluid” through a disordered medium. Here, the word fluid is quite generic: a liquid or a gas, or more generally, any phenomenon that spreads throughout a system that contains a degree of randomness or stochasticity. Examples include a fire spreading in a forest, electrons hopping between sites in composite materials, a disease spreading throughout a society, and so on. Generally speaking, the spread of a fluid through

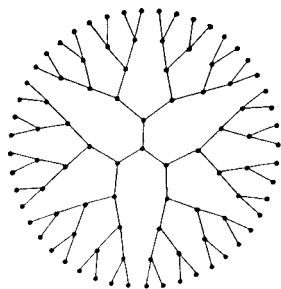


Figure 3.2 A Bethe lattice of coordination number 3.

- 2) *Paul John Flory* (1910–1985) made fundamental contributions to understanding of macromolecules and polymers in solution for which he received the Nobel Prize in Chemistry in 1974. He taught at Stanford and Carnegie Mellon Universities, and also worked in industry.
- 3) *Walter Hugo Stockmayer* (1914–2004) was an internationally renowned chemist who

made seminal contributions to polymer science. He taught at Dartmouth College and founded *Macromolecules*, the premier journal dedicated to polymers.

- 4) *John Michael Hammersley* (1920–2004) was a British mathematician who made important contributions to the theory of self-avoiding walks, percolation theory and Monte Carlo methods.

a medium may involve some stochasticity. One must realize, however, that the underlying mechanism of the stochasticity may be of two distinct types:

1. The stochasticity is dictated by the *fluid* in that it is the fluid that “decides” what path to take in order to spread in the system. This is the classical *diffusion process* that has been studied for 200 years.
2. The stochasticity in the fluid’s path is imposed by the *medium* through its morphology, which was the new phenomenon studied by Broadbent and Hammersley (1957). They named the phenomenon a *percolation process* because they thought that the spread of a fluid through a random medium resembles the flow of coffee in a percolator. Since we know that fluid and transport in a porous medium are influenced by the pore space morphology, they must be a kind of percolation process and, indeed, they are.

3.2.1

Bond and Site Percolation

As described above, we represent a porous medium by a network in which the bonds and sites represent, respectively, the throats and pores. In their original paper, Broadbent and Hammersley focused on two problems:

1. *Bond percolation* in which the bonds of a network are either occupied or *open* (to passage of a fluid) randomly and independently of each other with probability p , or they are vacant or *closed* (to passage of a fluid) with probability $1 - p$. For a large network, the probability p is equivalent to a random *fraction* p of all the bonds being open or intact. Figure 3.3 shows a square network in which a fraction p of the bonds are intact or open. For example, to model single-phase flow through a porous medium represented by its equivalent network, the open bonds may be thought of as the high-permeability regions of the pore space through which most of the fluid flows, whereas the vacant or closed bonds represent the very low-permeability (or nearly impermeable) regions of the pore space that contribute little, if any, to flow of the fluid. In natural porous media, the high- and low-permeability regions are not necessarily distributed randomly. However, for now, we ignore such complications. For flow of two immiscible fluids through a porous medium, the set of throats occupied by one fluid may be thought of as the open bonds, in which case the rest of the throats would represent the closed bonds (for now, we ignore the thin layers that a wetting fluid may form on the walls of the pores and throats occupied by the non-wetting fluid).

Two sites are called *connected* if there exists at least one path between them consisting solely of open bonds. A set of connected sites bounded by closed bonds is called a *cluster*. If the network is very large and p is sufficiently small, the size of any cluster is likely to be small. On the other hand, if p is close to one, the network should be almost entirely connected, apart from occasional small

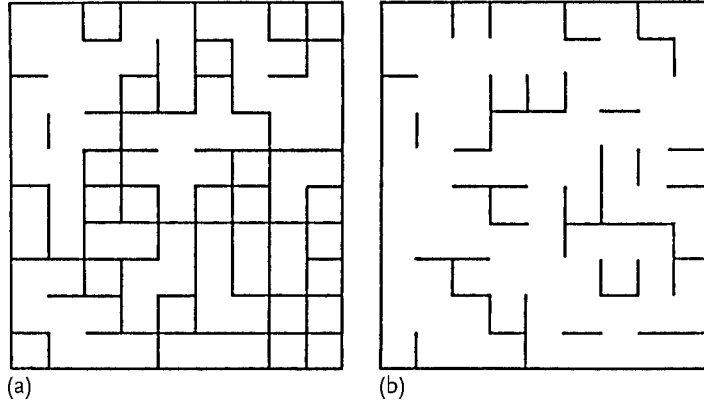


Figure 3.3 Bond percolation in the square network for $p = 1/3$ (a) and $p = 2/3$ (b).

“holes” formed by the closed bonds. Thus, there must be a well-defined value of p , which we denote by p_{cb} , at which a *transition* occurs in the macroscopic connectivity of the network: For $p_{cb} - \epsilon$, there is no *sample-spanning cluster* of open bonds that connects two opposing faces of the network, while for $p_{cb} + \epsilon$, such a cluster exists, where $\epsilon \rightarrow 0$. p_{cb} is called the *bond percolation threshold* of the network. Physically, p_{cb} is the *largest* fraction of the open bonds below which (at $p_{cb} - \epsilon$) there is no sample-spanning cluster of such bonds. Clearly, if there is no sample-spanning cluster of the open bonds, no macroscopic fluid flow through the network occurs. As we will see later in this book, bond percolation is relevant to describing single-phase flow in porous media with a broad distribution of pore sizes, and also certain two-phase flow problems.

2. *Site percolation* in which the sites of a network are occupied or open (to a fluid) with probability p and vacant or closed (to a fluid) with probability $1 - p$. Two nearest-neighbor sites are connected if they are both open, and clusters of such sites are defined in an obvious way. There is also a *site percolation threshold* p_{cs} above which (at $p_{cs} + \epsilon$) a sample-spanning cluster of occupied or open sites connects two opposing faces of the network. Figure 3.4 shows site percolation on a square network in which a fraction p of the sites are open.

Note that the percolation phenomenon as defined above is a *static* process. That is, once a percolation network with a given value of p is generated, its configuration does not change with time. Such static networks suffice for modeling certain flow phenomena in porous media. For other problems, for example, displacement of one fluid by another immiscible fluid in a porous medium, the configuration of the network in terms of the open and closed bonds and/or sites is dynamic. We will consider such dynamic percolation networks later in this book, although, it must be emphasized that many concepts described here are equally applicable to both static and dynamic percolation networks.

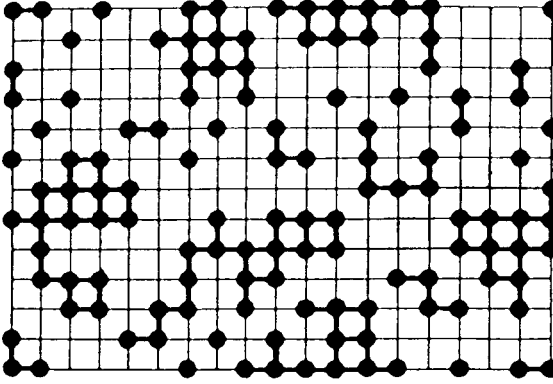


Figure 3.4 Site percolation in the square network for $p = 1/2$. Circles are the occupied sites.

The derivation of the exact values of p_{cb} and p_{cs} is an extremely difficult problem. In fact, exact derivation of these quantities has been possible only for networks related to the Bethe lattice for which

$$p_{cb} = p_{cs} = \frac{1}{Z - 1}, \quad (3.1)$$

where Z is the coordination number of the network, that is, the number of bonds connected to the same site, and for many two-dimensional (2D) networks. We compile the most accurate estimates of p_{cb} and p_{cs} (and their exact values if known) for some common 2D and 3D networks in Table 3.1.⁵⁾ Also shown, is the product $B_c = Z p_{cb}$ which, as can be seen, is an almost invariant of percolation networks,

Table 3.1 Percolation thresholds and $B_c = Z p_{cb}$ for some common 2D and 3D networks.

Network	Z	p_{cb}	B_c	p_{cs}
2D				
Honeycomb	3	$1 - 2 \sin(\pi/18) \simeq 0.6527^a$	1.96	0.6962
Square	4	$1/2^a$	2	0.5927
Triangular	6	$2 \sin(\pi/18) \simeq 0.3473^a$	2.08	$1/2^a$
3D				
Diamond	4	0.3886	1.55	0.4299
Simple-Cubic	6	0.2488	1.49	0.3116
BCC	8	0.1795	1.44	0.2464
FCC	12	0.119	1.43	0.199

^a Exact result.

5) The thresholds for many other networks are listed at en.wikipedia.org/wiki/Percolation_threshold (accessed 23 March 2010).

depending only on the dimensionality d of the network, with

$$B_c \simeq \frac{d}{d-1}. \quad (3.2)$$

The significance of B_c is discussed later in this chapter.

3.2.2

Computer Simulation and Counting the Clusters

Creating a percolating network by randomly removing sites or bonds is not actually suitable for engineering applications because such a procedure generates, in addition to the sample-spanning cluster, isolated finite clusters that have no significance to the problem studied. In most problems involving fluid flow through a network model of porous media, only the sample-spanning cluster is important. Alternatively, the phenomenon of interest may begin with a single cluster, but the dynamics of the phenomenon may lead later on to the formation of isolated clusters. An example is the displacement of an incompressible fluid by a second immiscible fluid. Therefore, if we randomly generate a percolation network, we must first delete all the isolated clusters. Alternatively, and much more efficiently, we may use a method due to Leath (1976) and Alexandrowicz (1980) that only generates the sample-spanning cluster for $p > p_c$, or the largest cluster for $p \leq p_c$, where p_c is either the bond or site percolation threshold.

Consider site percolation. One begins with a single open site at the network's center. The nearest-neighbor sites of the open site are then identified and considered occupied, and added to the cluster if random numbers R , uniformly distributed in $(0, 1)$ and attributed to the sites, are less than the given value p for which the cluster is to be generated. The *perimeter* – the nearest-neighbor empty sites – of the open sites are identified and the process of occupying the sites continues in the same way. If a selected perimeter site is not made occupied, then it remains unoccupied forever. The generalization of the method for generating clusters of occupied bonds is obvious, but is slightly more tedious.

An important task in computer simulation of percolation problems, including those related to fluid flow through porous media, is to count the number of clusters of a given size. For example, during displacement of an incompressible fluid A by an immiscible fluid B, one is interested in estimating the number of blobs of fluid A of a given size that are completely trapped by B, which is equivalent to computing the number of clusters of a given size in a percolation network. Hoshen and Kopelman (1976) developed an efficient algorithm for this task that is described in detail by Stauffer and Aharony (1994)⁶⁾, who also give a computer program for implementing the algorithm. Al-Futaisi and Patzek (2003) extended the algorithm to non-network systems.

6) Dietrich Stauffer (1943–), emeritus professor of physics at Cologne University in Germany, has made seminal contributions to percolation, phase transition, gelation, large-scale scientific computations, and the application of statistical physics to social and biological science.

3.2.3

Bicontinuous Porous Materials

An important question in modeling of two-phase flow in porous media is whether both fluid phases can *simultaneously* form sample-spanning clusters; that is, whether both fluids can be flowing. If this is possible, then the porous medium is called *bicontinuous*. For example, if the open and closed bonds or sites of a network model of a disordered porous medium contain two immiscible fluids A and B (say, oil and water), then percolation theory provides a clear answer to the question of bicontinuity of the porous medium. Random percolation possesses *phase-inversion symmetry*, that is, the morphology of that part of the system that contains fluid A at volume fraction p is *statistically identical* to that containing fluid B with the *same* volume fraction. Clearly, then, no 1D material can be bicontinuous, as its (bond or site) percolation threshold p_c is one. For spatial dimensions $d \geq 2$, any system possessing phase-inversion symmetry is bicontinuous for $p_c < p < 1 - p_c$ (where p_c is either the site or bond percolation threshold), provided that $p_c < 1/2$. Thus, all the 3D networks listed in Table 3.1 can be bicontinuous in either bond or site percolation. For 3D networks, however, neither $p_c < 1/2$ nor phase-inversion symmetry is a necessary condition for the bicontinuity.

Two-dimensional porous media are much more difficult to be made bicontinuous. In many respects, randomly-disordered two-phase materials correspond to site percolation systems. If so, since there is no 2D network with a site percolation threshold $p_{cs} < 1/2$ (see Table 3.1), we may conclude that no 2D randomly-disordered porous medium can be bicontinuous.

3.3

Connectivity and Clustering Properties

In addition to the percolation thresholds p_{cb} and p_{cs} , the behavior of percolation networks is quantified by several other important quantities. Some of such quantities describe the connectivity and clustering properties of the networks, while others characterize their effective flow and transport properties that we describe in the next section. In particular, some of the most important properties that describe connectivity and clustering of networks are as follows (p_c denotes p_{cs} or p_{cb}).

1. The *accessible fraction* $X^A(p)$ is that fraction of the occupied or open bonds (or sites) that belong to the sample-spanning cluster.
2. The *backbone fraction* $X^B(p)$ is the fraction of occupied or open bonds in the sample-spanning cluster that actually participate in a flow or transport process through the network since some of the bonds in the cluster are *dead-end*. In a flow experiment, the pressure drop across the dead-end bonds is zero. Therefore, $X^A(p) \geq X^B(p)$.
3. The *correlation length* $\xi_p(p)$ is the *typical* radius of percolation clusters for $p < p_c$, and the typical radius of the “holes” for $p > p_c$ that are generated by the

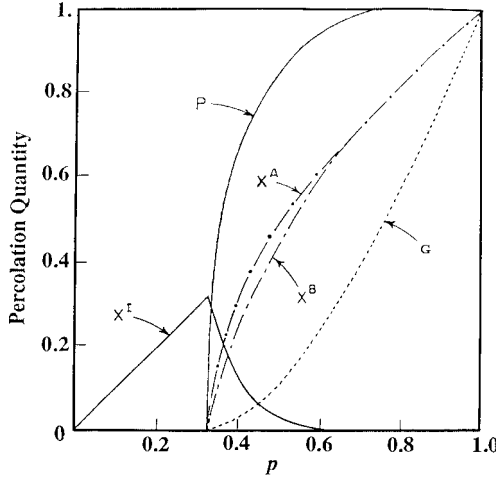


Figure 3.5 Typical p -dependence of the various percolation properties (site percolation in the simple-cubic network). $G = g_e$ is the effective conductivity, while $P(p)$ is the *percolation probability*, the probability that a site belongs to the sample-spanning cluster.

closed bonds or sites. Physically, the correlation length is very important in that it is, for $p > p_c$, the length scale over which the network is *macroscopically homogeneous*. Therefore, in computer simulation of fluid flow through a network model of porous media, (1) the linear size L of the network must be larger than ξ_p in order for the effective properties of the network, such as its permeability or diffusivity, to be independent of L , and (2) only for length scales that are larger than ξ_p can one use the classical continuum equations of flow and transport to describe such phenomena.

4. The *average number of clusters of size s* (per network site), $n_s(p)$, is important to the problem of two-phase flow in porous media because it corresponds, in a percolation model of the phenomenon, to the number of blobs (clusters of pores and throats) of a given size that are filled by an incompressible fluid and trapped by a second immiscible fluid. Figure 3.5 shows the typical p -dependence of these quantities in site percolation in the simple-cubic network.

3.4

Flow and Transport Properties

We must also consider the effective flow and transport properties of percolation networks as models of porous media, namely, their (brine-saturated) electrical conductivity, diffusivity, permeability, and other properties that will be studied throughout this book. Consider the problem of estimating the effective permeability of a porous medium represented by a network in which the pores' and throats' sizes

are broadly distributed. Under such conditions, a certain fraction of the bonds and sites (throats and pores) are too small to carry any significant amount of fluid and, therefore, can be removed from the network (porous medium). Doing so immediately generates a network in which a certain fraction of bonds and sites carry a fluid, while the rest of them are closed to the fluid. That is, the problem of computing the effective permeability of the porous medium is reduced to a percolation problem. Clearly, if the fraction of the bonds that are too small to carry any significant fluid is equal to or larger than the percolation threshold of the network, the effective permeability will be zero.

Consider a second example, namely, the problem of estimating the effective permeabilities to two immiscible and flowing fluids, A and B, in the same porous medium, or its corresponding network model, assuming that the spatial distribution of the two fluids throughout the network – random or otherwise – is known (see Chapters 14 and 15). Since the two fluids are immiscible, then, so far as computing the effective permeability to, that is, fluid A is concerned, one may cut all the bonds and sites (throats and pores) that are occupied by fluid B since fluid A cannot enter them. Doing so generates a percolation network in which some bonds (and sites) carry fluid A, while the rest of the bonds and sites are cut, or are closed to fluid A. Clearly, if the fraction of the bonds that are occupied by fluid A is smaller than or equal to the bond percolation threshold of the network, the effective permeability to fluid A is zero.

Similarly, computing the effective diffusivity D_e of a fluid in the same porous medium can also be recast in terms of a percolation problem. Hence, since a certain fraction of the bonds and sites are too small to accommodate any diffusing fluid, they can be removed from the network. How the effective permeabilities or diffusivity are computed by such network models will be described in Chapters 9 and 10.

3.5

The Sample-Spanning Cluster and Its Backbone

The sample-spanning cluster through which fluid flow and transport take place can be divided into two parts: The dead-end part that carries no flow and the backbone which is the multiply-connected part of the cluster. Near p_c , the bonds in the backbone are also divided into two groups: Those that are in the *blobs* that are multiply connected and make the flow and transport paths very tortuous, and the *red bonds* that are those that, if cut, would split the backbone into two parts. Such bonds are called red because in transport of heat or electrical currents through the network they carry the largest currents and, therefore, in analogy with a real electrical network, become very hot and thus red. Figure 3.6 shows a sample-spanning percolation cluster and its backbone.

Currently, the most efficient algorithm for identifying the backbone of a percolation network is due to Sheppard *et al.* (1999) (see also, Knackstedt *et al.*, 2000), which is an optimized algorithm for *simultaneous* identification of the minimal

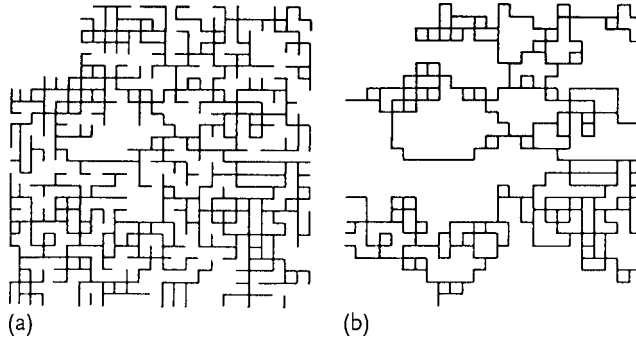


Figure 3.6 The sample-spanning cluster (a) and its backbone (b) in the square network for $p = 0.58$.

path length, the sites comprising the *elastic backbone*, and the usual backbone defined above. The minimal path is the set of the occupied or open sites (and the associated bonds) that are on the shortest path between two occupied and widely separated sites, while the elastic backbone is (Herrmann *et al.*, 1984) the set of the open sites that lie on the union of all the shortest paths between two widely separated open sites. There are three major steps in implementing the algorithm of Sheppard *et al.* (1999) that are as follows.

1. Using a breadth-first search algorithm, one labels each site in the cluster with its “cluster distance” from the inlet face, and then uses this information to burn backwards from the outlet face to identify the elastic backbone. At the same time, one constructs the “branch points list” – a list of all the cluster sites that are adjacent to the elastic backbone but are not part of it. The branch points list is ordered with the sites closest to the inlet face listed first. Note that the sites in the elastic backbone are also part of the usual backbone.
2. The algorithm stops if the branch points list is empty. Otherwise, one performs a depth-first search from the last site in the branch points list, flagging all the sites that are visited. During the search, unexplored branch points are added to their list, while another list tracks the sites that have been flagged as visited. One then performs an optimization during the depth-first search: If there are multiple branches from a single site, the site labeled as being closest to the inlet face is always the first to be explored.
3. The depth-first search terminates when one of two conditions are satisfied: (1) The search contacts the backbone again at a different site from which it started, in which case the sites in the visited-sites list are flagged as backbone sites, or (2) it retreats back to its starting site, at which point there will be no sites left in the visited-sites list. The algorithm continues at step 2.

3.6

Universal Properties

One of the most important characteristics of percolation networks is as follows. The behavior of many percolation quantities near p_c is insensitive to the microstructure (for example, the coordination number) of the network, and to whether the percolation process is a site or a bond problem. The quantitative statement of this *universality* is that many percolation properties follow *power laws* near p_c , and the *critical exponents* that characterize such power laws are universal and only depend on the Euclidean dimensionality d of the system. In general, the following power laws hold near p_c ,

$$X^A(p) \sim (p - p_c)^\beta, \quad (3.3)$$

$$X^B(p) \sim (p - p_c)^{\beta_{bb}}, \quad (3.4)$$

$$\xi_p(p) \sim |p - p_c|^{-\nu}. \quad (3.5)$$

For large clusters near p_c , the cluster size distribution $n_s(p)$ is described by the following scaling law,

$$n_s \sim s^{-\tau_p} f\left[(p - p_c)s_p^\sigma\right], \quad (3.6)$$

where τ_p and σ_p are two more universal critical exponents, and $f(x)$ is a scaling function such that $f(0)$ is not singular. For random percolation models, or for correlated percolation in which the correlations are short-ranged, all of the above critical exponents are completely universal.

Similar power laws are also followed by flow and transport properties of percolation networks. In particular, for the electrical conductivity g_e of the (brine-saturated) network and its permeability K_e ,

$$g_e(p) \sim (p - p_c)^{\mu_p}, \quad (3.7)$$

$$K_e(p) \sim (p - p_c)^\epsilon. \quad (3.8)$$

In most cases, $\mu_p = \epsilon$. The conditions under which $\mu_p \neq \epsilon$ will be described below. The power law that characterizes the behavior of the effective diffusivity $D_e(p)$ near p_c is derived from that of $g_e(p)$. According to Einstein's relation, $g_e \propto \rho D_e$, where ρ is the density of the carriers. Although a diffusing species (carrier) can move on all the clusters, above p_c , only diffusion on the sample-spanning cluster contributes significantly to D_e so that $\rho \propto X^A(p)$. Hence, $g_e(p) \propto X^A(p) D_e(p)$ and, therefore,

$$D_e(p) \sim (p - p_c)^{\mu_p - \beta}. \quad (3.9)$$

No exact relation is known between the flow, transport, and connectivity exponents. This is perhaps because the flow and transport exponents describe *dynamical* properties of percolation systems, whereas the connectivity exponents describe the *static* properties. The exponents μ_p and ϵ are mostly universal. In Table 3.2, the most accurate estimates of the critical exponents in 2D and 3D are compiled.

Table 3.2 The critical exponents (and fractal dimensions) of percolation. The connectivity exponents and the fractal dimension D_f for 2D systems as well as all the quantities for Bethe lattices are exact.

Exponent	$d = 2$	$d = 3$	Bethe lattice
β	5/36	0.41	1
β_{bb}	0.48	1.05	2
τ_p	187/91	2.18	5/2
σ_p	36/91	0.45	1/2
ν	4/3	0.88	1/2
D_f	91/48	2.53	4
D_{bb}	1.64	1.87	2
μ_p	1.3	2.0	3
e	1.3	2.0	3

3.7

The Significance of Power Laws

If two physical phenomena in disordered media, and in particular porous media, that contain percolation-type disorder (those in which the connectivity of the media's microscopic elements is important to their macroscopic properties) are described by two distinct sets of critical exponents, then the physical laws governing the two phenomena must be *fundamentally different*. Thus, the critical exponents help one to differentiate between different *classes* of problems and the physical laws that govern them. Moreover, since the numerical values of the percolation properties are not universal and vary from one system to another, but the scaling and power laws that they follow near p_c are universal and do not depend on the system's details, estimates of the critical exponents for a certain phenomenon are used for establishing the relevance of a particular percolation model to that phenomenon in disordered media. Finally, sometimes the power laws (3.7)–(3.9) are valid over a wider range than one expects. In that case, they are also very useful for *correlating* the data and interpreting them in terms of a well-understood theory, that is, percolation theory.

3.8

Dependence of Network Properties on Length Scale

As mentioned above, the correlation length ξ_p has the physical significance that for length scales $L > \xi_p$, the percolation system is macroscopically homogeneous. However, for $L < \xi_p$, the system is *not* homogeneous and its macroscopic properties depend on L . In this regime, the sample-spanning cluster is *statistically self-similar* at all the length scales less than ξ_p . Statistical self-similarity (see, for exam-

ple, Feder, 1988) implies that if we look at the cluster either at larger or smaller magnifications (scales), its structure looks the same and has the same statistical properties. In that case, the cluster's mass M (the total number of occupied bonds or sites that it contains) scales with L as

$$M \propto L^{D_f}, \quad L < \xi_p, \quad (3.10)$$

where D_f is the cluster's *fractal dimension* because the cluster's mass is not large enough to fill up the Euclidean space with dimension d . For $L > \xi_p$, one has $M \propto L^d$ and thus $D_f = d$, where d is the system's spatial dimension. The crossover between the fractal and Euclidean regimes takes place at $L \simeq \xi_p$. D_f is related to the critical exponents defined above. The mass M of the cluster is proportional to $X^A(p) \xi_p^d$ since only a fraction $X^A(p)$ of all the occupied bonds or sites are in the cluster. Therefore, we must have $M \propto \xi_p^{d-\beta/\nu}$. For $L < \xi_p$, we must replace ξ_p with L since in this regime, L is the dominant (and the only relevant) length scale. Thus, $M \propto L^{d-\beta/\nu}$, which, when compared with power-law (3.10), yields

$$D_f = d - \frac{\beta}{\nu}. \quad (3.11)$$

Similarly, for $L < \xi_p$, the backbone is also a fractal object and its fractal dimension D_{bb} is given by

$$D_{bb} = d - \frac{\beta_{bb}}{\nu}. \quad (3.12)$$

Note that at $p = p_c$, the correlation length is divergent so that at this point, the sample-spanning cluster and its backbone are fractal objects at *any* length scale. Table 3.2 also lists values of D_f and D_{bb} .

If $L < \xi_p$, then all the properties of the network become scale dependent. Since $|p - p_c| \sim \xi_p^{-1/\nu}$, one can rewrite the power laws (3.3)–(3.5) and (3.7)–(3.9) in terms of ξ_p . For example, power law (3.3) is rewritten as $X^A(p) \sim \xi_p^{-\beta/\nu}$. As before, for $L < \xi_p$, one replaces ξ_p by L and, therefore,

$$X^A(L) \sim L^{-\frac{\beta}{\nu}}, \quad (3.13)$$

$$X^B(L) \sim L^{-\frac{\beta_{bb}}{\nu}}, \quad (3.14)$$

$$g_e(L) \sim L^{-\frac{\mu_p}{\nu}}, \quad (3.15)$$

$$k_e(L) \sim L^{-\frac{e}{\nu}}, \quad (3.16)$$

$$D_e(L) \sim L^{-\theta}, \quad (3.17)$$

where $\theta = (\mu_p - \beta)/\nu$.

Scale-dependent properties are usually a signature of a fractal morphology. Therefore, the question is, what is the significance of fractal morphology? The

correlations between the various parts of a fractal system are *long-ranged*. In fact, if a system has a fractal structure at any length scale (for example, the sample-spanning cluster at p_c), then the correlations exist everywhere at that scale. Flow and transport in such systems *cannot* be described by the classical continuum equations. We will come back to this very important point later in this book.

3.9

Finite-Size Effects

All the percolation properties described so far, including the percolation thresholds and the critical exponents, are defined for those that are of infinite extent. However, percolation in finite systems is important since, both in practical applications and in computer simulations, one usually works with systems of finite extent. In such systems, as p_c is approached, the correlation length ξ_p eventually exceeds the system's linear size L , in which case, as pointed out above, L becomes the dominant length scale. Fisher (1971) developed a theory for the scaling properties of a finite thermodynamic system near a critical temperature, usually called *finite-size scaling*. Adopting Fisher's theory for percolation systems implies that *any* property P_L of a system of linear size L is written as

$$P_L \sim L^{-\zeta} f(u) \quad (3.18)$$

with $u = L^{1/\nu}(p - p_c) \sim (L/\xi_p)^{1/\nu}$, where $f(u)$ is a non-singular function. If, in the limit $L \rightarrow \infty$, P_∞ follows a power law, such as, $P_\infty \sim (p - p_c)^\delta$, then one must have $\zeta = \delta/\nu$. Therefore, the variations with L of $P_L(p)$ in a *finite network* at the percolation threshold of the *infinite network* can be used to obtain information about the quantities of interest for an *infinite network near* p_c , where power laws (3.3)–(3.9) hold.

Finite-size scaling theory has been successfully used for obtaining accurate estimates of the critical exponents and even the percolation thresholds from simulation of finite systems. The finite size of a network causes a shift in its percolation threshold (Levinshtein *et al.*, 1976):

$$p_c - p_c(L) \sim L^{-\frac{1}{\nu}}. \quad (3.19)$$

In this equation, p_c is the percolation threshold of the infinite system and $p_c(L)$ is an *effective* p_c for a finite system of linear dimension L . However, we should point out that Eqs. (3.18) and (3.19) are valid for very large network size L , whereas in practice, very large systems cannot be easily simulated. To remedy this situation, Eq. (3.18) is modified to

$$P_L \sim L^{-\zeta} [a_1 + a_2 h_1(L) + a_3 h_2(L) + \dots], \quad (3.20)$$

where a_1 , a_2 and a_3 are three fitting parameters, and h_1 and h_2 are called the *correction-to-scaling* functions that are particularly important when small and moderate system sizes L are used in the simulations. For flow and *transport properties*,

$h_1 = (\ln L)^{-1}$ and $h_2 = L^{-1}$ often provide accurate estimates of ζ (Sahimi and Arbabi, 1991). Equation (3.20) provides us with a means of estimating a critical exponent: Calculate P_L at $p = p_c$ for several system sizes L and fit the results to Eq. (3.20) to estimate ζ and, thus, $\delta = \zeta \nu$.

3.10

Random Networks and Continuum Models

Although percolation in regular networks – those in which the coordination number Z is the same everywhere – has been extensively invoked for studying flow and transport in disordered porous media, percolation in continua and in topologically-random networks – those in which the coordination number varies from site to site – are of great interest since, in almost any practical situation, one must work with such irregular and continuous systems. For example, percolation in continuous systems is directly applicable to the characterization and modeling of morphology and effective transport properties of many types of heterogeneous materials (Sahimi, 2003a), although network models that take into account the effect of the fine details of the morphology of such materials are equally applicable and, in fact, easier to use. There are at least three ways of realizing a percolating continuum:

1. One inserts a random (or correlated) distribution of inclusions, for example, circles, spheres or ellipses, in an otherwise uniform background. The correlations arise when one imposes certain constraints on the system. For example, if the particles are not allowed to overlap, or if the extent of their penetrability or overlap is fixed, then the resulting morphology contains correlations with an extent that depends on the type of the constraints imposed. In such models, percolation is defined either as the formation of a sample-spanning cluster of the channels *between* the inclusions, or as the formation of a sample-spanning cluster of touching or overlapping inclusions.
2. The percolating system is generated by tessellating the space into regular or random polyhedra (Winterfeld *et al.*, 1981). A (volume) fraction of the polygons (polyhedra) are designated as one phase of the material, while the rest constitutes the second phase, with each phase having its own distinct effective properties. Figure 3.7 presents an example of such a model which is called the *Voronoi tessellation*. In this model, random (Poisson) points are distributed in a 2D system. The Voronoi polygons are then constructed around each point with the condition that every point inside a polygon is closer to its Poisson point than those of the neighboring polygons. The extension of the method to 3D is conceptually straightforward.
3. The third method consists of constructing a distribution of sticks of a given aspect ratio or plates of a given extent inserted in a uniform background. Such a model has been used for representing the fracture network of rock, with the porous matrix represented by the background, and will be studied in detail in Chapter 8.

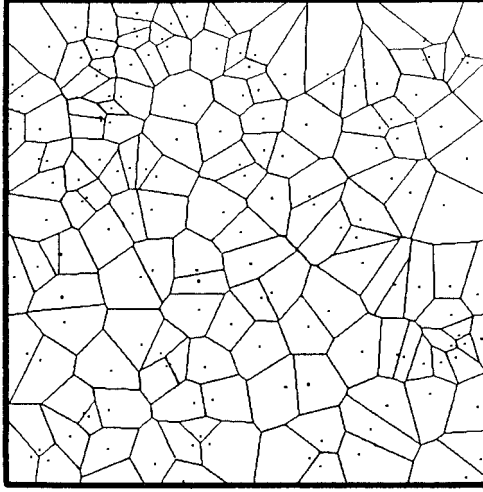


Figure 3.7 Two-dimensional Voronoi tessellation. The Voronoi network is obtained by connecting the centers of the neighboring polygons.

Somewhat similar to percolation on networks described so far, the percolation thresholds of continuum d -dimensional systems with $d \geq 2$ are not exactly known, except for certain 2D symmetric-cell models. Two-phase symmetric-cell models are constructed by partitioning space into cells of arbitrary shapes and sizes, with the cells being randomly designated as phases 1 and 2 with probabilities p_1 and p_2 . A simple example is a 2D system that is tessellated into square cells, where each cell belongs either to phase 1 or phase 2 with probabilities p_1 and p_2 . The Voronoi tessellation provides another example of such models. The symmetric-cell models possess phase-inversion symmetry defined in Section 3.2.3.

It is generally believed that for any 2D symmetric-cell model in which the cells' centers are the sites of a fully triangulated lattice, one has $p_{1c} = p_{2c} = 1/2$, although we are not aware of any rigorous proof of this prediction. In this case, the continuum percolation threshold and the site percolation threshold of the associated network are identical. Examples include hexagonal and Voronoi tessellations, both of which have site percolation thresholds of $1/2$.

Suppose that particles that are distributed in a uniform matrix constitute phase 2 of a continuum percolation model of a disordered material (such models are used for studying porous media; see Chapter 7), with the background matrix being phase 1. Pike and Seager (1974) and Haan and Zwanzig (1977) were among the first to obtain accurate estimates of the percolation threshold p_{2c} for randomly distribution of overlapping disks and spheres. The most accurate estimate of p_{2c} for 2D (disk) systems currently available is due to Quintanilla *et al.* (2000) who reported that $p_{2c} \simeq 0.67637$, while for 3D (spherical particles) systems, Rintoul and Torquato (1997) reported $p_{2c} \simeq 0.2895$. The corresponding values for overlapping oriented squares and cubes (Pike and Seager, 1974; Haan and Zwanzig, 1977) are $p_{2c} \simeq 0.67 \pm 0.01$ and 0.28 ± 0.01 , respectively. In these models, all the particles

had the same size. In general, if the particles do not all have the same size, then p_{2c} should continuously depend on the size distribution of the particles. Numerical simulations indicate (see, for example, Pike and Seager, 1974; Lorenz *et al.*, 1993), however, that p_{2c} only weakly depends on the particle size distribution. Meester *et al.* (1994) conjectured that the monodisperse distribution *minimizes* the percolation threshold.

One of the most important discoveries for continuum percolation (Scher and Zallen, 1970) is that a critical occupied volume fraction ϕ_c , which is defined as

$$\phi_{2c} = p_{cs} f_1, \quad (3.21)$$

where f_1 is the filling factor of a network when each site of the network is occupied by a sphere in such a way that two nearest-neighbor impermeable spheres touch one another at the midpoint, appears to be an almost invariant of the system with a value of about 0.17 for 3D systems. Shante and Kirkpatrick (1971) generalized this idea to permeable spheres and showed that the average number B_c of bonds per sites at p_c is related to ϕ_c by

$$\phi_{2c} = 1 - \exp\left(-\frac{B_c}{8}\right), \quad (3.22)$$

and that for continuous systems, $B_c \rightarrow p_{cs} Z$ for large Z . It is clear from Table 3.1 and Eq. (3.2) that in 3D, $B_c \simeq 1.5$. It has been shown (see, for example, Balberg and Bienbaum, 1985) that the connectivity exponents defined by power laws (3.3)–(3.6) are equal for networks and continuous systems.

Similar to network models described earlier, one may also speak of two-phase continuum models of porous media that are bicontinuous. Hence, extending to continuum systems the criteria of bicontinuity for networks that was described above, we say that a d -dimensional two-phase continuum that possesses phase-inversion symmetry is bicontinuous for $p_{2c} < p_2 < 1 - p_{2c}$ for $d \geq 2$. However, these are not necessary conditions. For example, a bicontinuous structure without phase-inversion symmetry but with $p_{2c} < 1/2$ is a 3D distribution of identical overlapping spheres which has been used for decades for modeling various disordered media, where for $0.29 < p_2 < 0.97$, both the particle phase and the pores between the particles percolate. Another example of a bicontinuous system without phase-inversion symmetry and with $p_{2c} < 1/2$ is a close-packed face-centered-cubic network, used as a model of packed beds and unconsolidated porous media (see Chapters 7, 9, 14, and 15). For example, at the percolation threshold of the particle (sphere) phase, $p_{2c} = \pi/\sqrt{18}$, the space between the particles also percolates.

3.11

Differences between Network and Continuum Models

Certain properties of flow and transport in percolating continua can be quite different from that in the network models. Consider, for example, a model in which

one inserts at random circular or spherical inclusions in an otherwise homogeneous medium. For the obvious reason, this is called the *Swiss-cheese model* (see Figure 3.8). If fluid flow takes place through the matrix, then the problem is mapped onto an equivalent problem on the *edges* of the Voronoi polygons or polyhedra (Kerstein, 1983), a network that has an exact coordination number of four in 3D. One may also construct the *inverted Swiss-cheese model* in which the roles of the two phases in the Swiss-cheese model are switched, that is, a transport process such as conduction takes place through the circular or spherical inclusions (a good model of heat transfer through packed beds), in which case the transport problem can be mapped onto an equivalent one in the Voronoi *network* which is constructed by connecting to each other the Poisson points in the neighboring polygons or polyhedra (see Figure 3.7).

It is assumed in both models that all the inclusions have the same size. Van der Marck (1996) suggested a more general method for mapping transport in continua that consists of a distribution of inclusions of various sizes in a uniform background onto an equivalent problem in an equivalent network model. We should, however, point out that if one utilizes the equivalent network of a given continuum, one can no longer assign the flow and transport properties of the network's bonds from an *arbitrary* distribution because there is a *natural* distribution of the properties of the flow and transport channels in the continuum which must be constructed based on the shapes and sizes that the channels take on (see below).

The Voronoi network was utilized by Jerauld *et al.* (1984b,d) and Sahimi and Tsotsis (1997) as a prototype of irregular networks to study transport in disordered materials. The *average* coordination number of the Voronoi network is about 6 and 15.5 in 2D and 3D, respectively. Jerauld *et al.* (1984b,d) established that as long as the average coordination number of a regular network and a topologically-random one are about the same, many flow and transport properties of the two systems are, for all practical purposes, identical. For example, the 2D Voronoi network has a bond-averaged mean coordination number of about 6.3, while the coordination number of the triangular network is exactly 6.

However, certain continuous percolation models violate the universality of the flow and transport exponents (Halperin *et al.*, 1985; Feng *et al.*, 1987) described

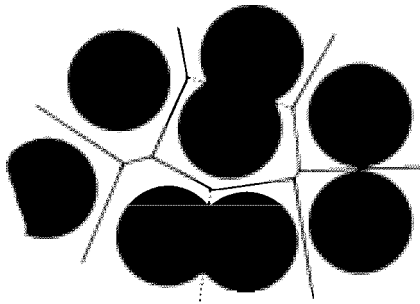


Figure 3.8 The 2D Swiss-cheese model.

above, in which case the flow and transport exponents for the continuous models are not necessarily the same as those in random networks and, thus, they must be estimated separately. The differences between the critical exponents of lattice and continuum percolation are caused by the natural distributions of the flow and transport properties that give rise to new power laws that cannot be predicted by the network models, unless the same natural distributions are also utilized in the network models as well (Halperin *et al.*, 1985; Feng *et al.*, 1987).

3.12

Porous Materials with Low Percolation Thresholds

Many natural porous media have very small percolation thresholds. That is, their permeability, diffusivity and electrical conductivity are nonzero even for very small values of porosity, the volume fraction of their void space. One may also prepare porous materials that have exceedingly small percolation thresholds. This can be achieved if the particle size distribution is very broad, and each particle possesses a “soft” repulsive interparticle potential with a range that is larger than the size of the particle. Unlike monodisperse particles, polydispersity causes the particles to fill the space, but the repulsive interactions prevent formation of a sample-spanning cluster until the system is essentially completely filled by the particles. An example of this type of material is colloidal dispersions. The percolation threshold of such materials, that is, the porosity at which a fluid can flow through them, is exceedingly low.

3.13

Network Models with Correlations

In the network and percolation models described thus far, no correlations existed between various segments of the system. However, disorder in many porous materials is not completely random. There are usually correlations with an extent that may be finite but large. For example, in packing of solid particles, there are short-range correlations.

The earliest correlated percolation model that we are aware of is a correlated bond percolation model due to Kirkpatrick (1973). In his model, to each site of a network is assigned a random number s_i , uniformly distributed between -1 and $+1$. Each bond ij is assigned a number b_{ij} which is calculated by

$$b_{ij} = \frac{1}{2} (|s_i| + |s_j| + |s_i - s_j|) , \quad (3.23)$$

and all the bonds with $b_{ij} > \epsilon$ are removed, where ϵ is some selected value. Thus, a bond remains intact only if b_{ij} is sufficiently small, implying that $|s_i|$ and $|s_j|$ must both be small. Therefore, if a bond is present or open, its neighbors are also likely to be open and, hence, the open bonds are clustered together. Many other per-

colation models with short-range correlations have been developed, most of which are described elsewhere (Sahimi, 1998).

Positive correlations (that is, correlations that cluster the open bonds together, as opposed to negative correlations that make it more likely that an open bond is next to a closed one, and vice versa), whether short- or long-ranged, usually *reduce* the percolation threshold of the network. For example, in Kirkpatrick's model, $p_{cb} \simeq 0.1$ for the simple-cubic network, much lower than $p_{cb} \simeq 0.2488$ for random bond percolation (see Table 3.1). This is due to the clustering of the open bonds caused by the positive correlations, as a result of which formation of sample-spanning flow or transport paths is possible even at low values of the fraction of the open bonds.

Interesting percolation models with long-range correlations in which the correlations decay with the distance between two points on the sample-spanning cluster, or even increase, have also been developed. Most of such models have been described by Sahimi (2003a). Some are relevant to fluid flow and transport in field-scale porous media, and will be described later in Chapters 10 and 11.

3.14

A Glance at History

Before closing this chapter, it may be interesting to give a brief review of the history of the application of percolation theory to modeling porous media problems. Despite the fact that Broadbent and Hammersley had expressed the hope that their theory would someday be used for solving some practical problems involving porous media, explicit use of concepts of percolation theory for describing flow phenomena in porous media gained popularity only in the 1980s. Since “who was the first to use percolation” has been a matter of some contention and controversy, it may be useful to review the history to see “who said what and when”, at least according to the published papers in the open literature.

To our knowledge, Torelli and Scheidegger (1972) were the first who recognized the relevance of percolation theory for modeling fluid flow and transport in porous media. They were interested in hydrodynamic dispersion in porous media (see Chapters 11 and 12) and pointed out that percolation theory, if appropriately modified and applied, may provide useful insight into its properties. They did not, however, use percolation and, in fact, they did not even report any results in their paper.

Melrose and Brandner (1974) suggested that the entrapment of oil in reservoir rock is similar to percolation processes, and proposed that an approach based on percolation may yield deeper insight into the problem, but they did not actually calculate anything. Davis *et al.* (1975), who studied transport processes in composite media, remarked at the end of their paper that, “Although, to our knowledge, no quantitative work has been done on the subject, we believe that 2-phase oil-water flow in oil fields is a percolation process in which the connectivity of each phase determines the relative permeability of that phase.” However, they also did not report any results.

Larson, Scriven, and Davis (1977)^{7,8)} also suggested that percolation theory may be useful for describing entrapment of one fluid phase by another in porous media. To demonstrate the usefulness of their idea, they calculated the percolation cluster size distribution n_s in a Bethe lattice for various coordination numbers and made a qualitative comparison between the results and the relevant experimental data. Almost simultaneously, Chatzis and Dullien (1977) calculated several percolation properties of various 2D and 3D networks, and pointed out how they may be used for simulating two-phase flow in porous media. They compared their predictions with the measured capillary pressure curves. Levine *et al.* (1977) discussed the application of percolation theory to wetting/dewetting phenomena in porous media, and pointed out how the effective-medium approximation (see Chapters 9, 8, and 12) may be used for estimating the permeability of a porous medium. Shortly after the three papers, de Gennes and Guyon (1978)⁹⁾ also suggested that two-phase flow problems in porous media may belong to the class of percolation processes. They used visualization of mercury porosimetry as an example, and proposed ideas for using percolation concepts for modeling it and other processes in porous media.

Finally, two papers in 1980 further established the applicability of percolation for modeling two-phase flow in porous media. Lenormand and Bories (1980) proposed a percolation model, now popularly known as invasion percolation (see Chapter 15), for modeling a drainage process, that is, a process in which a non-wetting fluid displaces a wetting fluid from a porous medium. Golden (1980) discussed the application of percolation theory for studying two-phase flow problems and the associated hysteresis (history-dependent) phenomena that are routinely observed in porous media.

After publication of these original papers, there was an explosion of new ideas and methods for modeling porous media problems using percolation theory. We shall review these concepts and methods in the appropriate chapters of this book.

- 7) *L. Edward “Skip” Scriven* (1931–2007) was Regents Professor of Chemical Engineering and Materials Science at the University of Minnesota. A member of the National Academy of Engineering, he made seminal contributions to coating flows, flow through porous media, interfacial phenomena, and complex fluids.
- 8) *Howard Theodore “Ted” Davis* (1937–2009) was Regents Professor of Chemical Engineering and Materials Science and former Dean of the Institute of Technology at the University of Minnesota. A member

of the National Academy of Engineering, he made fundamental contributions to statistical mechanics of surfaces and interfaces, flow through porous media, and complex fluids.

- 9) *Pierre-Gilles de Gennes* (1932–2007) was a French physicist who made fundamental contributions to superconductivity, liquid crystals, polymers physics, and wetting. A professor at Orsay and College de France, he was director of École Supérieure de Physique et de Chimie Industrielles de la Ville de Paris (ESPCI). Among other awards, he received the Nobel Prize for Physics in 1991.

4

Characterization of the Morphology of Porous Media

Introduction

In this chapter, we describe various morphological properties of a porous medium as well as the experimental techniques that have been used over the past several decades for characterizing the morphology. A porous medium morphology consists of (1) the geometry that describes the pores' shapes and sizes and the structure of their internal surface, and (2) the topology that quantifies the way pores and throats are connected together. We restrict our discussions to characterization of *laboratory-scale* porous media. Chapter 5 will describe characterization of *field-scale* porous media, for which one needs to utilize geostatistical techniques.

To comprehend the morphology of a porous medium, one must have an understanding of how the medium was originally formed. While man-made porous materials usually have well-understood mechanisms of formation, the same is not necessarily true about natural porous media. The morphology of rock is directly linked to the *diagenetic processes* that lead to its formation. Such processes begin with deposition of sediments, followed by compaction and alteration processes that cause drastic changes in the rock's morphology. Consider, for example, sandstone that is an assemblage of grains with a wide variety of chemical compositions. If the environment around sandstone changes, its grains begin to react and produce new compounds, which also change the sandstone's mechanical properties. The chemical and physical changes in the sand after the deposition constitute the diagenetic processes, the main features of which are, (1) mechanical deformation of grains; (2) solution of grain minerals; (3) alteration of grains, and (4) precipitation of pore-filling minerals, cements, and other materials. The latter three features involve changes in the chemical composition of rock, which are usually induced by transport of some reactants in the pore space. These phenomena are called *metasomatic processes* and their influence is key to the content of rock.

Diagenesis starts immediately after deposition; it continues during burial and uplift of rock until outcrop weathering reduces it again to sediment. Such changes produce an end product with specific diagenetic features, the nature of which depends on the initial mineralogical composition of rock as well as the composition of the surrounding basin-fill sediments. Given a porous medium with a particu-

lar mineralogical composition, its diagenetic history depends on several factors, including time-dependent exposure to varying temperature and pressure, and the chemistry of the pore fluid. Together, such factors constitute the *historical aspects* of rock.

The diagenetic processes lead to a variety of rock morphology. Pores can take on essentially any shape or size, and may also be highly interconnected. Patsoules and Cripps (1983) used scanning electron microscopy to study rock and to obtain information about the pores' shapes, sizes, and connectivity, and the roughness of their surface. They reported that the rock that they examined, which was upper cretaceous chalk from East Yorkshire (in England) and the North Sea, contained highly interconnected pores. Some of the ring-shaped pores of the chalk were connected to at least 25–30 other pores, and remained connected even when the porosity was very low. Therefore, one important effect of the diagenetic process is to keep the pore space highly interconnected. The rock alteration processes involve complex phenomena, for example, nucleation on the pores' surface and mineral crystal growth – time-dependent phenomena that reduce rock's porosity and permeability. Reducing the sample's permeability also reduces the flow rate, implying that the rate of nucleation of mineral crystals increases. The crystals cannot, however, grow indefinitely because they are limited by the growth rate at the time they are nucleated. Moreover, the growth of new mineral crystals inhibits that of the older ones. It is the competition between nucleation of new mineral crystals and the growth of the older crystals that determines the distribution of the crystal sizes.

Classical experimental methods for characterization of a laboratory-scale porous medium include mercury porosimetry and sorption measurements. However, the proper interpretation of the data is not straightforward and requires careful modeling. It has also become possible over the past decade to use tomographic methods, for example, microfocus X-ray (MFX) imaging (Liu and Miller, 1999), magnetic resonance imaging (MRI) (Manz *et al.*, 1999), and three-dimensional (3D) transmission electron microscopy (TEM) in order to examine the pore space of various types of porous media. Such methods are, however, not general enough to be used for a wide variety of porous materials. Although 3D TEM methods can be used (Koster *et al.*, 2000) to study morphologies with pores as small as 1–30 nm, the samples' thickness should be 500 nm at most, much smaller than those of laboratory-scale rock samples or even porous catalysts. Techniques that are based on nuclear MRI can also be used (see, for example, Hollewand and Gladden, 1995) to study macroscopic (larger than 10 μm) variations in the microscopic properties of porous media. Such techniques are, however, relatively expensive and not used as widely as mercury porosimetry and sorption measurements, which remain to be the “workhorse” techniques of characterization of porous media.

Before starting our discussion, let us point out that according to the International Union of Pure and Applied Chemistry, the pores of any porous medium should be classified based on their sizes, which we will adopt throughout this book:

1. *Micropores* (or *nanopores*) with sizes ≤ 2 nm.
2. *Mesopores* with sizes in the range 2–50 nm.
3. *Macropores* with sizes ≥ 50 nm.

4.1

Porosity

The porosity of a porous medium is the volume fraction of its voids. While the origin of porosity in man-made porous materials is usually known and understood, the same is not necessarily true about natural porous media. For example, the porosity of rock has either a *primary* or a *secondary* origin. The former is due to the original pore space of the sediment, whereas the latter is caused by unstable grains or cements undergoing chemical and physical changes through reaction with the formation water, and have partially or entirely passed into the solution. Therefore, if the pore space is restored through dissolution of authigenic minerals, then the original porosity that had been protected from precipitation by deposition of minerals is converted into secondary porosity. According to Schmidt and McDonald (1979), solution pores provide more than half of all the pore space in many sedimentary rocks.

The significance of the secondary porosity in carbonate rock has been recognized for a long time, but its importance to sandstones has only relatively recently been appreciated (Hayes, 1979). As discussed by Schmidt and McDonald (1979), there are five classes of secondary porosity in sandstones, defined according to their origin: (1) fracturing; (2) shrinkage; (3) dissolution of sedimentary grains and matrix; (4) dissolution of authigenic pore-filling cement, and (5) dissolution of authigenic replacive minerals.

Five different kinds of pores may contain secondary porosity, namely, (1) intergranular pores; (2) oversized pores; (3) moldic pores; (4) intraconstituent pores, and (5) open fractures. Of these, fractures are distinctly different from the other four types of pores and, therefore, will be considered in Chapters 6 and 8. The existence of the secondary porosity can sometimes be recognized even with the naked eye. Other indications of the existence of secondary porosity include oversized or elongated pores, corroded and fractured grains, and several others.

In Chapter 3, where we described percolation properties of network models of disordered media, we distinguished p , the fraction of the open bonds or sites (throats or pores) of the network, regardless of whether they can be reached from two opposing external surface of the network, from the accessible fraction $X^A(p)$ of the open bonds or sites, which is the fraction of the open bonds or sites that are in the sample-spanning cluster that connects the two opposing surfaces. Clearly, we always have $X^A(p) \leq p$. Likewise, we must distinguish the *total* porosity ϕ of a porous medium from the *accessible* porosity ϕ_A which is the volume fraction of that part of the void space that can be reached from its external surface. In this sense, p and $X^A(p)$ are the analogs of ϕ and $\phi_A(\phi)$. Therefore, we may also define a *critical porosity* ϕ_c – the analog of the percolation threshold p_c – such that for

$\phi \leq \phi_c$, there would be no sample-spanning cluster of voids that connect two opposing surfaces of a porous medium, whereas for $\phi > \phi_c$, the sample-spanning cluster does exist, and flow and transport through the porous medium can occur.

Porosity may be measured by several methods. The simplest of such techniques is a direct method in which the medium's total volume V_t is measured. The porous medium is then crushed to remove all the void space, and the volume V_s of the resulting solid is measured. The total porosity is then given by $\phi = 1 - V_s/V_t$. Clearly, this method measures a porous medium's total porosity. Another method of estimating ϕ is by inspecting thin sections of porous media under a microscope. The area fraction of the pores seen in the thin sections is an estimate of the porous medium's total ϕ . One may also estimate ϕ by a stochastic method, due to Chalkley *et al.* (1949), using a photomicrograph. In this method, a pin is thrown N times into the picture. A *hit list* is compiled whereby whenever the pin crosses any void area in the picture, a hit is registered. If the experiment is repeated for a large enough N , then, $\phi = N_h/N$ where N_h is the total number of hits.

One of the most widely used methods for measuring the accessible porosity is the so-called *gas expansion* method in which the porous medium is enclosed in a container filled with a gas such as air. Clearly, the gas only penetrates the accessible void space of the porous medium. The container is then connected to a second evacuated container which causes a change in its pressure. The accessible porosity of the system is then estimated from

$$\phi_A = 1 - \frac{V_1}{V_s} - \frac{V_2}{V_s} \frac{P_f}{P_f - P_i}, \quad (4.1)$$

where V_1 is the volume of the container in which the porous system is enclosed, V_2 the volume of the evacuated container, V_s is the volume of the porous sample, and P_i and P_f are, respectively, the initial and final pressures of the medium. Several other methods of measuring ϕ and ϕ_A are described by Collins (1961) and Scheidegger (1974). Table 4.1 lists the total porosity of several classes of porous materials.

Table 4.1 Porosity ranges for several classes of porous media.

Porous medium	ϕ (%)
Black slate powder	57–66
Silica powder	37–49
Random packing of spheres	36–43
Sand	37–50
Sandstone	8–38
Limestone (dolomite)	4–10
Coal	2–12
Concrete	2–7

Let us point out that Hilfer (1992) proposed a local porosity theory that has proven to be a very useful property for characterization of porous media and the development accurate models for them. The main idea is to measure porosity (or other well-defined observables) within a bounded compact subset of the porous medium and to prepare histograms of the data to be used for analysis and modeling. The local porosity concept is far superior to the concept of pore size distribution that, as discussed below, is often vague and imprecise. One can also define local percolation probabilities (see Chapter 3) that characterize the local connectivity properties of a sample porous medium. We will come back to these concepts in Chapter 7.

4.2

Fluid Saturation

The saturation S of a fluid is the volume fraction of the void space filled by that fluid. Similar to porosity, fluid saturation can be measured by several methods. For example, one may weigh a porous sample of known porosity before and after it is filled by a fluid, from which the fluid saturation is estimated. If a porous material does not conduct electricity and is partially filled by a conducting fluid (such as salt), then by measuring the resistivity of the partially-saturated porous sample, the fluid saturation is estimated based on Archie's law (Archie, 1942):

$$R_e = R_0 S^{-n}, \quad (4.2)$$

where R_e and R_0 are, respectively, the resistivity of the partially-saturated porous medium, and that of the fluid, and n is called the Archie exponent. For clean sands, $n \simeq 2$, however, in general, n can be less than or greater than two. If the porous medium contains chemically-active chemical compounds, for example, clays and shales, Archie's law must be modified.

The spatial distribution of a fluid's saturation in a porous medium – a dynamic property that evolves with the time if the fluid is accompanied by other immiscible fluids in the medium – is also of particular significance. Many techniques have been suggested for mapping out the saturation distribution, ranging from gamma- and X-ray absorption (Boyer *et al.*, 1947; Laird and Putnam, 1959) and microwave transmission techniques (Aggarwal and Johnstone, 1986), to computerized tomography using X-ray and techniques based on nuclear magnetic resonance (NMR) (Baldwin and Yamanashi, 1986; Mandava *et al.*, 1990; Chen *et al.*, 1993, 1994; Liaw *et al.*, 1996). In addition, ultrasonic methods have been proposed (Soucemarianadin *et al.*, 1989) that are based on the difference between the sound velocities in various liquids saturating a porous medium.

Among these, the NMR technique has received considerable attention, and has gained traction as a viable and accurate method. Submillimeter resolution or better can be obtained. The technique is based on the fact that the intrinsic magnetization intensity is proportional to the amount of the observed fluid phase within a voxel in an image of the medium. The relatively fast relaxation that is associated with fluids

in porous media, however, causes the attenuation of the intensity by an amount that depends on the characteristic relaxation of the fluids as they reside within the porous media and the particular pulse sequence used. Determination of the saturation distribution directly leads to the determination of the porosity distribution as well.

4.3

Specific Surface Area

An important property of a porous medium is its specific surface area Ξ , defined as the ratio of the internal surface area of the voids and the bulk volume of the porous medium that, therefore, is expressed as a reciprocal length. Ξ can be measured by several techniques (Scheidegger, 1974). For example, one can use a photomicrograph of polished sections of a sample porous medium with sufficient contrast between the pores and the matrix. From the relation between 2D (surface) measurements and the properties of the 3D system, an estimate of Ξ is obtained. Sorption experiments may also be used for measuring Ξ (see Section 4.13), although they depend on the size of the probe molecules and, therefore, may underestimate Ξ . It is clear that, similar to porosity, one can distinguish between the *accessible* specific surface area, which is measured by a probe based on the surface area of the accessible voids of a porous medium, and the *total* specific surface area.

4.4

The Tortuosity Factor

A third characteristic of a porous medium is its tortuosity τ , which is usually defined as the ratio of the true or total length L_t of the diffusion path of a fluid particle diffusing in the porous medium, and the straight-line distance L between the starting and finishing points of the particle's diffusion, $\tau = L_t/L$ that, by definition, is always greater than (or at least equal to) one. Clearly, τ depends on the porosity of a porous medium. It should also depend on the molecular size of the diffusing particles. For $\phi \approx \phi_c$, where ϕ_c is the critical porosity or the percolation threshold, τ is very large. At ϕ_c the tortuosity *diverges* as $L_t \rightarrow \infty$ at ϕ_c . In the classical continuum models of flow and transport in porous media, τ is often treated as an adjustable parameter.

Since the tortuosity factor is defined in terms of the true path length of diffusing particles, it is sometimes expressed in terms of the particles' diffusivity. If D_e and D_0 are, respectively, the effective diffusivities of diffusing probes in a porous medium and in the bulk (outside the porous medium), then,

$$D_e = \frac{\phi_A D_0}{\tau} . \quad (4.3)$$

Often, ϕ instead of ϕ_A is used in Eq. (4.3).

4.5

Correlations in Porosity and Pore Sizes

In most models of flow and transport in porous media, spatial correlations in pore sizes and porosity are either neglected, or assumed to have a limited extent. However, it has been suggested that *long-range* or extended correlations are likely to exist in many, if not all, natural porous formations both at the pore (Knackstedt *et al.*, 1998) and field scales (see Chapter 5). Characterizing the correlations in the pore space of complex porous media requires the ability to examine the microstructure of their pore space. For years, direct measurements of the pore-space characteristics were largely restricted to the stereological studies of thin sections of porous media (Pathak *et al.*, 1982; Doyen, 1988; Ghassemzadeh and Sahimi, 2004a,b). However, thin sectioning requires a considerable amount of time to polish, slice, and digitize the sample. As mentioned in the introduction, modern imaging techniques now allow one to observe highly complex morphologies in 3D in a minimal amount of time. In particular, X-ray computed tomography (CT) is a nondestructive technique for visualizing features in the interior of opaque solid objects and for resolving information on their 3D geometries. Conventional CT can be used to obtain the porosity map of a piece of a porous medium at length scales down to a millimeter (see, for example, Hicks *et al.*, 1992). High-resolution CT (see, for example, Spanne *et al.*, 1994) has made it possible to measure the geometric properties at length scales as small as a few microns.

Millimeter-scale CT images of Berea sandstone were presented by Knackstedt *et al.* (2001b) (see Figure 4.1). Heterogeneity in the porosity distribution is evident even by visual inspection. Analysis of the porosity distribution shown in Figure 4.1 revealed (Knackstedt *et al.*, 2001b) that the extent of the spatial correlations in the

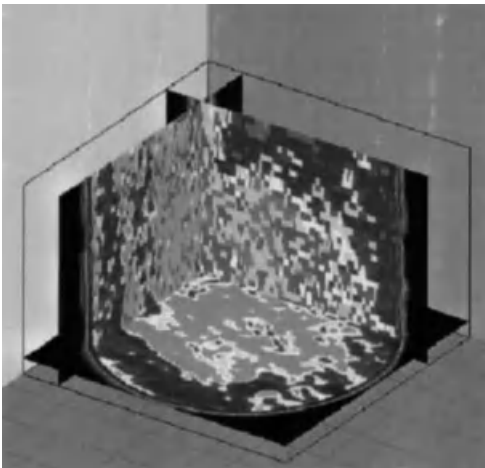


Figure 4.1 Gray-scale image of the porosity distribution in a sandstone at 1 mm pixel resolution. The clustering of the low- and high-porosity areas is visible, implying correlations (after Knackstedt *et al.*, 2001b).

distribution was about 3 mm. The variance in the porosity distribution is described by a stochastic process called the fractional Brownian motion (FBM) that induces extended correlations in the value of the stochastic variable. We will show in Chapter 5 that the same stochastic process also describes the spatial correlations in the porosity and permeability distributions of many field-scale porous media, for example, oil reservoirs. Chapter 6 will show that the same stochastic function describes the roughness of the internal surface of fractures. More heterogeneous sandstones exhibit correlated heterogeneity over a more extended range, on the order of 1 cm or more. Other data on carbonate rock revealed spatial correlations in porosity on the order of 5 mm (Xu *et al.*, 1999). The correlation length measured from such CT images pertains to porosity (pore clustering) rather than a direct measure of correlations in the pore sizes, but the same type of correlations also exist in the pore sizes.

Micro-X-ray CT image facilities can now provide 1024^3 voxel images of porous materials at a voxel resolution of less than $6\text{ }\mu\text{m}$ (Spanne *et al.*, 1994). Knackstedt *et al.* (2001b) obtained a $512 \times 512 \times 666$ image of crossbedded sandstone at 10 micron resolution via micro-CT imaging. In Figure 4.2, we compare two consecutive series of six sections of the crossbedded sandstone. The two series of images are separated by less than *one* millimeter. One can see a large change in the porosity with such a small change in the depth. Pore and throat sizes, and other geometric properties of the rock differ significantly, despite the images only being two grain diameters apart. Figure 4.3 shows a trace of 660 values of the porosity measured at a separation of $10\text{ }\mu\text{m}$, indicating again the FBM-type correlations, which has also been found to describe correlated heterogeneities of rock at the *meter* scale through borehole analysis (see, for example, Makse *et al.*, 1996a,b). We will come back to this point in Chapter 5 where we describe the properties of field-scale porous media. Direct measurement of pore and throat sizes, the correlations between them, and also between neighboring pore volumes were also made on the crossbedded sandstone and on four samples of Fontainebleau sandstone (Lindquist and Venkatarangan, 1999; Lindquist *et al.*, 2000). The results indicated that there is strong correlation between the volume of a throat and the average volume of the pores to which they are connected. Thus, direct measurements of pore-scale structure strongly question the common assumption that rock properties at the pore scale are randomly distributed.

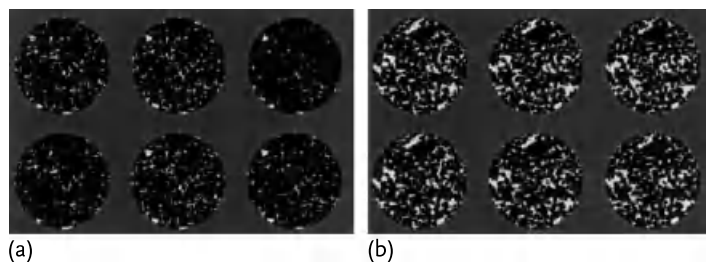


Figure 4.2 Comparison of two sets of consecutive slices of a crossbedded sandstone at $10\text{ }\mu\text{m}$ spacing. (a) $\phi < 0.1$; (b) less than 1 mm away, $\phi > 0.15$ (after Knackstedt *et al.*, 2001b).

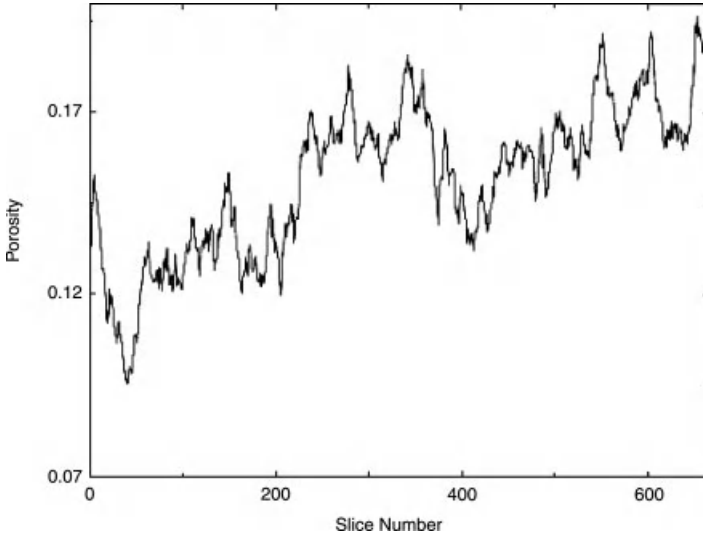


Figure 4.3 Variations of the porosity of the slices of the sandstone shown in Figures 4.1 and 4.2 (after Knackstedt *et al.*, 2001b).

4.6

Surface Energy and Surface Tension

If gravitational and other forces are absent or can be neglected, a liquid assumes a spherical shape because, among all the geometries, it possesses the minimum surface-to-volume ratio. Suppose that the sphere is distorted by an external force that increases the surface area. Then, liquid's molecules must migrate to the surface in order to provide the surface area. To do so requires expending some work W in order to raise the potential energy of a molecule since the number of stabilizing interactions between the molecule and its neighbors decreases once the molecule reaches the surface. The free energy G of the liquid increases by the work W and, therefore, the change ΔG in the free energy represents the *free surface energy* G_s . This implies that ΔG_s , the change in the free surface energy, represents the net work required to alter the surface area of a material. Spontaneous processes are usually associated with a decrease in free energy which is why in the absence of external forces, liquids spontaneously take on a spherical shape in order to minimize their exposed surface area and, hence, their free surface energy. This also explains why when two droplets of similar fluids are brought into contact, they spontaneously form a larger drop.

The surface tension σ of a substance is identical to the free surface energy G_s per unit area, $G_s = A\sigma$, where A is the surface area. In differential form,

$$\sigma = \left(\frac{\partial G_s}{\partial A} \right)_{T,V,n}, \quad (4.4)$$

where T is the temperature, V the total volume, and n the number of molecules.

4.7

Laplace Pressure and the Young–Laplace Equation

According to the discussion in Section 4.6, a bubble should collapse in order to minimize its free surface energy. As the shrinking process proceeds, however, the pressure of the gas inside the bubble increases and counters the reduction in the bubble's radius. If the radius of the bubble decreases from r to $r - \Delta r$, the free energy decreases by $-\Delta G = 8\pi r \Delta r \sigma$, and the corresponding volume change is $4\pi r^2 \Delta r$. The shrinking compresses the gas inside the bubble, while the air outside the bubble expands. The net work ΔW associated with the compression-expansion is given by $-\Delta W = (P_i - P_o)(4\pi r^2 \Delta r)$, where P_i and P_o are, respectively, the pressure inside and outside the bubble. Because the net work should be equal to the change in the free energy, one obtains

$$P_i - P_o = \Delta P = \frac{2\sigma}{r}. \quad (4.5)$$

ΔP is sometimes referred to as the Laplace¹⁾ pressure.

Equation (4.5) is a special limit of a more general equation known as the *Young²⁾–Laplace equation* (Laplace, 1806; Young, 1855), which generalizes Eq. (4.5) to non-spherical surfaces. If we consider an area element on a nonspherical curved surface, then there can exist two *principal radii of curvature*,³⁾ which are denoted by r_1 and r_2 . Suppose that r_1 and r_2 are held constant while a surface element is stretched from x to $x + dx$, and from y to $y + dy$, for which the work W_1 is given by $W_1 = [(x + dx)(y + dy) - xy]\sigma \simeq (x dy + y dx)\sigma$, where we have ignored the products of the differentials. At the same time, the area element is stretched due to an increase, $\Delta P = P_i - P_o$, in the pressure, which generates a displacement dz along the z -axis and a corresponding work W_2 given by $W_2 = [(x + dx)(y + dy)dz]\Delta P \simeq (xy dz)\Delta P$. However, since the two radii of curvature, r_1 and r_2 , remain unchanged, we must have $(x + dx)/(r_1 + dz) = x/r_1$, and, $(y + dy)/(r_2 + dz) = y/r_2$, which imply that $dx = x dz/r_1$ and $dy = y dz/r_2$. By substituting for dx and dy into the equation for W_1 and recognizing that at mechanical equilibrium, $W_1 = W_2$, we obtain

$$\Delta P = \left(\frac{1}{r_1} + \frac{1}{r_2} \right) \sigma, \quad (4.6)$$

1) *Pierre-Simon, marquis de Laplace* (1749–1827) made pivotal contributions to mathematical astronomy, statistics and other branches of science. He formulated the Laplace equation and Laplace transform, and was probably the first who contemplated the existence of black holes.

2) *Thomas Young* (1773–1829) was a British scientist who made important contributions to wave theory of light, solid mechanics (Young's modulus is named after him), and energy, and was also a prominent Egyptologist.

3) Suppose that the equations, $X = X(t)$, $Y = Y(t)$, and $Z = Z(t)$, define a curve in space as the parameter t varies over a specific range, and that s represents arc length along the curve. Then, the quantity r defined by

$$r^{-1} = \sqrt{\left(\frac{d^2 X}{ds^2}\right)^2 + \left(\frac{d^2 Y}{ds^2}\right)^2 + \left(\frac{d^2 Z}{ds^2}\right)^2}$$

is called the *radius of curvature*. Note that,

$$ds = \sqrt{(dX)^2 + (dY)^2 + (dZ)^2}.$$

which is the Young–Laplace equation. For a sphere, $r_1 = r_2$ and Eq. (4.6) reduces to Eq. (4.5).

4.8

Contact Angles and Wetting: The Young–Dupré Equation

Imagine that a drop of liquid is at rest on a solid surface. Then, the circumference of the contact area of a circular drop is drawn toward the drop's center by σ_{sl} , the solid–liquid interfacial tension. The (equilibrium) vapor pressure of the liquid generates an adsorbed layer on the solid surface that causes the circumference to move away from the drop's center. This generates a solid–vapor surface (or interfacial) tension, σ_{sv} . At the same time, the interfacial tension σ_{lv} between the liquid and vapor is equivalent to the liquid's surface tension σ , and acts tangentially to the *contact angle* θ – the angle at which the interface between the two fluids intersects the solid surface – and draws the liquid toward the center of the drop. Then, a force balance yields

$$\sigma_{sv} = \sigma_{sl} + \sigma_{lv} \cos \theta . \quad (4.7)$$

Equation (4.7) is known as the *Young–Dupré equation* (Young, 1855; Dupré, 1869), and is used to determine the contact angle:

$$\cos \theta = \frac{\sigma_{sv} - \sigma_{sl}}{\sigma_{lv}} . \quad (4.8)$$

If $\theta < 65^\circ$, then we say that the surface is *wetted* by the fluid because the *adhesive* forces exceed *cohesive* forces. In this case, the liquid spreads spontaneously on the surface. For $105^\circ < \theta < 180^\circ$, the surface is not wetted by the liquid because in this case, the cohesive forces are larger than the adhesive forces, and the liquid in the form of a drop remains stationary on the surface and takes on a pseudo-spherical shape. In between, for $65^\circ < \theta < 105^\circ$, the surface is said to be *intermediately-wet*. Note that Eq. (4.7) can also be derived by considering the amount of work ΔW done by moving the line of contact by a distance dx , $\Delta W = (\sigma_{sv} - \sigma_{sl})dx - (\sigma_{lv} \cos \theta)dx$. Since the work ΔW vanishes at equilibrium, we obtain the Young–Dupré equation by setting $\Delta W = 0$.

Such a description is also applicable to two liquids (as opposed to a liquid and its vapor described above). Thus, consider a situation in which a drop of, for example, water is placed on a surface immersed in oil. Then, the interface between the two fluids intersects the solid surface at a contact angle θ (see Figure 4.4). Associated with this system are three surface tensions corresponding to the two fluid–solid interfaces, and the water–oil interface. The surface tensions are related to each other through the Young–Dupré equation:

$$\sigma_{so} = \sigma_{sw} + \sigma_{ow} \cos \theta , \quad (4.9)$$

where σ_{so} and σ_{sw} are the surface tension between oil and the solid surface, and between water and the solid surface, respectively. It should then be clear that for

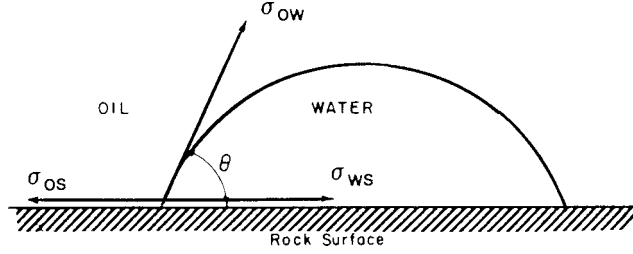


Figure 4.4 The contact angle formed between a pair of liquids (oil and water) and the pore surface.

$\theta < 65^\circ$, the surface is water-wet, for $\theta > 105^\circ$, it is oil-wet, while in between it is intermediately-wet. *Complete* wetting of a surface occurs for $\theta = 0^\circ$, and *total* non-wetting at $\theta = 180^\circ$. From Eq. (4.8), we obtain the adhesive forces F_A and cohesive forces F_C as

$$F_A = \frac{\sigma_{sv}}{\sigma_{lv}}, \quad F_C = \frac{\sigma_{sl}}{\sigma_{lv}}. \quad (4.10)$$

The quantity (or its equivalent for a liquid and its vapor),

$$C_s = \sigma_{so} - (\sigma_{sw} + \sigma_{ow}), \quad (4.11)$$

is called the *spreading coefficient*. One must have $C_s > 0$ for total wetting, while for partial wetting, $C_s < 0$. There are other ways of quantifying the wettability of a system, which will be described in Chapters 14 and 15.

4.9

The Washburn Equation and Capillary Pressure

Suppose that a capillary tube of radius r is immersed in a liquid. Then, the liquid will rise in the tube if the contact angle $\theta > 90^\circ$, or will be depressed below the surface of the liquid if $\theta < 90^\circ$. Suppose that the net height of capillary rise or depression is h . Consider, first, the case in which the liquid rises in the tube. Suppose that point A is in the gas phase next to the interface in the tube, and that point B is in the liquid in the tube at the same level as the free surface of the liquid outside the tube (thus, A and B are separated by a distance h). Clearly, the pressure P_A at A is given by $P_A = P_B + hg(\rho_l - \rho_g)$, where ρ_l and ρ_g are the densities of the liquid and the gas, and g is the gravitational constant. Therefore, $\Delta P = P_A - P_B = hg(\rho_l - \rho_g)$, which, in view of Eq. (4.5), implies that

$$hg(\rho_l - \rho_g) = \frac{2\sigma}{r}, \quad (4.12)$$

which is valid when the liquid completely wets the surface, $\theta = 0^\circ$. For $\theta > 0$, we proceed as follows. The work required for moving up the capillary tube is given by

$W = (\sigma_{sl} - \sigma_{sv})\Delta A = -(\sigma_{lv} \cos \theta)\Delta A$, where ΔA is the area of the tube's wall covered by the liquid, and we used Eq. (4.7). The same amount of work is needed to force out a column of height h out of the tube. Suppose that a volume V of the liquid is forced out of the tube with gas at a constant pressure ΔP_g above ambient. Then, $W = V\Delta P_g$. For a capillary tube of radius r and length ℓ , $V = \pi r^2 \ell$ and $A = 2\pi \ell$,

$$\Delta P = -\frac{2\sigma}{r} \cos \theta , \quad (4.13)$$

which was first presented by Washburn (1921).

An important quantity for characterization of a porous medium is the *capillary pressure* P_c . Suppose that two immiscible fluids, for example, water and oil, are in contact in a bounded system, for example, a cylindrical tube, and are separated by an interface. Then, there is a discontinuity in the pressure field as one moves across the interface from one fluid phase to the other. That is, if we consider two points, one in the water phase at pressure P_w and one in the oil phase at pressure P_o , both in the immediate vicinity of the interface, then, $P_w \neq P_o$. The pressure difference $P_c = P_o - P_w$ is called the capillary pressure (the pressure is usually higher in the non-wetting fluid, oil in this case), and is given by the Young–Laplace equation, Eq. (4.6), modified by the contact angle θ :

$$P_c = \sigma_{ow} \left(\frac{1}{r_1} + \frac{1}{r_2} \right) \cos \theta . \quad (4.14)$$

For an interface in a cylindrical tube $r_1 = r_2 = r$, where r is the radius of the tube. Note that the principal radii of curvature are measured from the non-wetting fluid side.

If we consider the capillary pressure in a simple system, for example, a cylindrical or a conic tube, then it is clear that P_c should depend *uniquely* on the amount of the fluid in the system and, hence, on its saturation. However, the situation is different in a porous medium with many irregularly-shaped pores because different saturations may yield the *same* P_c , and vice versa since there are several interfaces in the porous medium at different locations. This is indeed what one finds in an experiment measuring P_c in a porous medium. For example, suppose that we initially fill a porous medium with water, and then expel it gradually by injecting oil into the pore space. Suppose also that the water is the wetting fluid. Thus, injecting oil into the porous medium requires applying a pressure which must be increased as more oil enters the porous medium. At each stage of the experiment, and for each value of the water saturation S_w , there is a corresponding capillary pressure P_c . If we continue the experiment for a long enough time, we will reach a point where no more water is produced, although there may still be some water left in the pore space. At this point, that is, at the *highest* value of P_c , the water saturation is called the *irreducible* water saturation S_{iw} , and the process of expelling water by oil is called *drainage*.

If we now start with the same porous medium at the end of drainage (at the highest P_c) and expel the oil gradually by injecting water into the medium – a

process called *imbibition* –, we obtain another capillary pressure curve which is very different from what we obtained during drainage and, thus, there is *hysteresis* in the capillary pressure-saturation curves. Figure 4.5 shows a typical capillary pressure diagram during both drainage and imbibition. As can be seen, even when $P_c = 0$, there is still some oil left in the medium. The saturation of oil at $P_c = 0$ is called the *residual* oil saturation, S_{or} . Clearly, drainage or imbibition does not have to start at $S_w = 1$ or at $S_w = S_{iw}$, but can begin at any saturation in between the two, that is, we can have *secondary* drainage, secondary imbibition, and so on. If we carry out such experiments, we again find different capillary pressure curves, which is also shown in Figure 4.5.

Equation (4.14) can be rewritten in a more general form in order to take into account the effect of the pore geometry:

$$P_c = \frac{2\sigma}{r_t} G(\theta, p), \quad (4.15)$$

where p represents a set of parameters that describe the shape of the pore. For example, in order to take into account the fact that what is usually called a pore has a converging segment (the throat) and a diverging part (the pore that is connected to the throat's mouth), a pore shape has been used with a radius that is a sinusoidal function of the axial position (Oh and Slattery, 1979). In this case,

$$G(\theta, p) = \frac{\cos \theta + \left(\frac{A^* \pi}{L^*} \right) \sin \theta \sin \left(\frac{2\pi z^*}{L^*} \right)}{\left\{ 1 + \frac{A^*}{2} \left[1 - \cos \left(\frac{2\pi z^*}{L^*} \right) \right] \right\} \left[1 + \left(\frac{A^* \pi}{L^*} \right)^2 \sin^2 \left(\frac{2\pi z^*}{L^*} \right) \right]^{\frac{1}{2}}}. \quad (4.16)$$

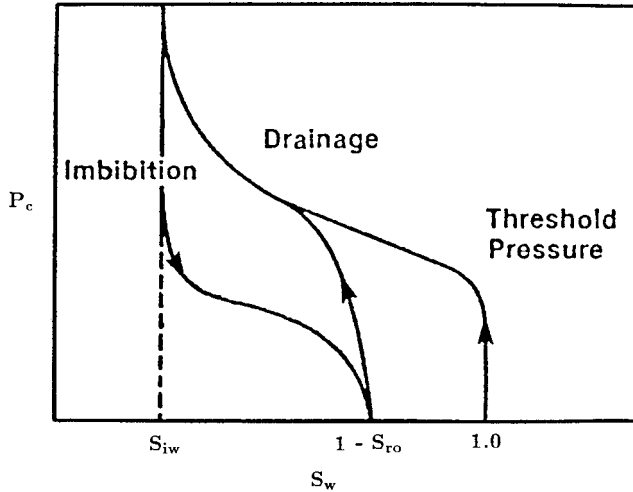


Figure 4.5 Typical capillary pressure curves for water-wet porous media.

Here, A is the amplitude of the sinusoidal function, L the pore's total length, z the axial position, and $*$ denotes a quantity made dimensionless by dividing it by r_t , the radius of the pore neck (the minimum pore radius). The expressions for capillary pressures in a variety of other pore geometries are given by Mason and Morrow (1984).

4.10

Measurement of Capillary Pressure

Capillary pressure curves are measured by several methods. However, such measurements are usually time consuming, as P_c is supposed to be an equilibrium property of a porous medium, and achieving true equilibrium is usually difficult and may take a long time.

One technique for measuring the capillary pressure is the centrifuge method in which a small sample is saturated by a wetting fluid and is placed in a container filled by a non-wetting fluid. The entire system is then rotated at an angular velocity ω . The density of the wetting fluid ρ_w is usually larger than that of the non-wetting fluid ρ_{nw} and, thus, the wetting fluid leaves the system at the outer radius. At the same time, the wetting fluid is replaced by the non-wetting fluid at the inner radius. Within the sample at an axial position z , one has $dP_w/dz = \rho_w g$, and, $dP_{nw}/dz = \rho_{nw} g$, and, therefore,

$$P_c(z) = P_c(z_0) + g \int_{z_0}^z (\rho_{nw} - \rho_w) dz, \quad (4.17)$$

where z_0 is a reference point. The capillary pressure at the inner radius R_1 of rotation of the sample is given by

$$P_c = \frac{1}{2} \omega^2 (\rho_w - \rho_{nw}) (R_2^2 - R_1^2), \quad (4.18)$$

where R_2 is the outer radius of rotation of the sample where the pressure has the same value in both phases, that is, where $P_c = 0$ (since water leaves the system there). For every ω , a different amount of water is expelled from the system and measured and, therefore, the capillary pressure–saturation diagram is constructed.

Measuring the saturations is complicated by the fact that, compared with the radius of the rotation, the dimensions of the sample porous medium are often not negligible. This implies that the centrifugal forces are not constant within the sample, as a result of which the saturation is not uniform within the sample. Therefore, the saturation used in the P_c diagram is an *average* value defined by

$$\langle S_w \rangle = \frac{1}{R_2 - R_1} \int_{R_1}^{R_2} S_w dr. \quad (4.19)$$

Equation (4.19) is often replaced by an approximate equation due to Hassler and Brunner (1945) and Slobod *et al.* (1951):

$$\langle S_w \rangle = \frac{1}{P_{c1}} \int_0^{P_{c1}} S_w(P_c) dP_c . \quad (4.20)$$

Upon differentiating Eq. (4.20), we obtain

$$\langle S_w \rangle + P_{c1} \frac{d\langle S_w \rangle}{dP_{c1}} = S_w(P_{c1}) , \quad (4.21)$$

which is used for constructing a saturation–capillary pressure curve.

The third method of measuring the capillary pressure is by mercury injection in which the non-wetting fluid is mercury while the wetting “phase” is the vacuum in the sample. We will come back to this method shortly, where we describe how mercury injection is used for determining the pore size distribution of a porous sample. In addition, in Chapter 14, we will describe capillary pressure for various types of porous media.

4.11

Pore Size Distribution

Every book and most research articles on porous media mention the “pore size distribution” of the pore space, but it is often not clear what is meant by the distribution. In an unconsolidated porous medium consisting of particles, the space between the particles is called the voids, whereas if the particles themselves are porous, then the void space in the particles is called the pores. However, careful examination of natural porous media reveals that what are usually referred to as the pores can, in fact, be divided into two groups. In the first group are the pore bodies where most of the porosity resides, while in the second group are the pore throats – the narrow channels that connect the pore bodies. One usually assigns *effective* radii to pore bodies and throats that, in reality, are nothing but the radii of spheres that have the same volume. Thus, pore bodies and pore throats are defined in terms of approximate maxima and minima of the largest-inscribed-sphere radius. As described in Chapter 3, in a network representation of a pore space, the pore bodies are represented by the network’s sites or nodes, while the pore throats are represented by its bonds. The volume of a pore body can be assigned to the corresponding node; alternatively, it can be apportioned among the network bonds, which is what is done in network modeling of flow and transport processes in porous media. Clearly, if the pore size distribution is known, then an average pore size, which is just the first moment of the distribution, can also be defined. However, a popular but empirical method of characterizing the variations in the pores’ and throats’ sizes is through a *mean hydraulic diameter* d_H , defined as

$$d_H = 4 \frac{V_p}{S_p} , \quad (4.22)$$

where V_p and S_p are the volume and surface of the pores, respectively. In view of the discovery that the pore volume and surface of many porous media are very rough and may have certain properties that are described by stochastic geometry (see below), d_H may not be such a useful quantity as it was thought of before the discovery because in the presence of surface roughness, d_H may depend on the length scale.

The pore size distribution is defined as follows: It is the *probability density function* that yields the distribution of pore volume by an effective or characteristic pore size. However, even this definition is somewhat vague because if the pores could be separated, then each of them could be assigned an effective size, in which case the pore size distribution would become analogous to the particle size distribution. However, because the pores are interconnected, the volume that one assigns to a pore can be dependent upon both the experimental method and the model of pore space that one employs to interpret the data. Four methods of measuring pore size distributions are mercury porosimetry, sorption experiments, small-angle scattering, and nuclear magnetic relaxation methods. The first two methods have been used extensively, while the latter two are newer and may, under certain conditions, be more accurate.

Let us point out that a method of measuring the pore size distribution, often used for porous membranes but applicable to other types of porous media, is based on flow permporometry. Detailed discussion of the method and a percolation model for interpreting the data and extracting the pore size distribution is given by Mourhatch *et al.* (2010).

4.12

Mercury Porosimetry

Mercury porosimetry as a probe of the structure of a porous material was first developed by Ritter and Drake (1945) and has remained popular ever since. It is usually used for pores between 3 nm and 100 μm . In this method, the porous medium is first evacuated and then immersed in mercury. Since mercury does not wet the pores' surface, it enters the pore space only if a pressure is applied. The pressure is then increased, either incrementally or continuously, and the volume of the injected mercury is measured as a function of the applied pressure. With mercury being a non-wetting fluid, its injection into a porous medium corresponds to drainage. However, the injection is usually called *mercury intrusion*. One needs to apply increasingly larger pressures in order for the mercury to progressively penetrate smaller pores. Very high pressures can even damage the internal structure of the medium, but we ignore them here. Clearly, if the porous medium consists of a packing of porous particles, mercury will first penetrate the voids between the particles and then, at higher applied pressures, the small pores within the particles. Given a particular porous sample, there is a *unique* characteristic maximum pressure associated with it, which is the pressure required to completely saturate the sample with mercury.

The pressure is then lowered back to ambient, as a result of which the mercury is retracted from the pores. With vacuum acting as the wetting phase, *mercury extrusion* or retraction corresponds to an imbibition process. The shapes of the intrusion–extrusion curves widely vary, as they depend on the morphology of the porous medium that one examines by mercury porosimetry.

Complete intrusion–extrusion experiments are sometimes called the *scanning loops*, defined as full intrusion–extrusion experiments that bring the system back to ambient pressure, but where the highest pressure achieved in the experiments is less than the characteristic (highest) pressure of the sample mentioned above. As a result, some of the pore space remains unfilled with mercury. One may also form *miniloops* that are somewhat different from the scanning loops. An intrusion miniloop consists of injecting mercury into the sample up to a particular pressure, followed by a partial retraction of the mercury (not necessarily down to ambient pressure), and then a re-intrusion back up to the given ultimate pressure. An extrusion miniloop consists of a primary intrusion, followed by a partial primary retraction which is then followed by a partial re-intrusion and then a secondary re-extrusion of mercury back to the pressure at the end of the original partial primary extrusion. Mini- and scanning loops were originally introduced by Reverberi *et al.* (1966) and Svata (1972).

An important discovery was made by Wardlaw and co-workers (Wardlaw and Taylor, 1976; Wardlaw and McKellar, 1981). They constructed a plot of cumulative residual saturation (the amount of mercury entrapped) versus initial saturation (corresponding to the highest pressure in the cycle) from scanning loop data, and found that by adding the cumulative residual saturation curve to the initial intrusion curve, a curve equivalent to a final re-intrusion (re-injection) curve is produced. The resulting re-injection curve is the one that results from increasing the pressure back up to the characteristic maximum pressure (see above), starting from ambient pressure, with the sample being in the state at the end of the primary retraction curve. Moreover, Wardlaw and McKellar (1981) suggested that by subtracting the initial intrusion curve from the respective re-injection curve, one would obtain a plot of residual saturation versus initial saturation without the need to conduct the full set of scanning-loop experiments. These empirical results were then proven to be true for any porous model (Androustopoulos and Salmas, 2000, where references to two earlier papers by the same authors are also given).

A precise apparatus for measuring mercury intrusion–extrusion curves was described by Thompson *et al.* (1987a,b), which consists of four components: (1) a mercury reservoir positioned on an elevator raised by a stepper-motor-driven screw; (2) a sample holder on a pan balance connected to the reservoir by stainless-steel tubing; (3) stainless-steel electrodes located on the top and bottom of the cylindrical sample, and (4) electronics for measurement of the AC resistance, the temperature, and the atmospheric pressure. The apparatus is shown in Figure 4.6. The experiments are automated by computer control. Before mercury injection is started, the pore space is evacuated to a pressure of 10^{-3} Pa. During the measurements, the elevator height is typically changed by 0.1–10 mm and the sample weight is monitored until equilibrium is reached. Typical experimental sensitivities are 10^{-5} cm³

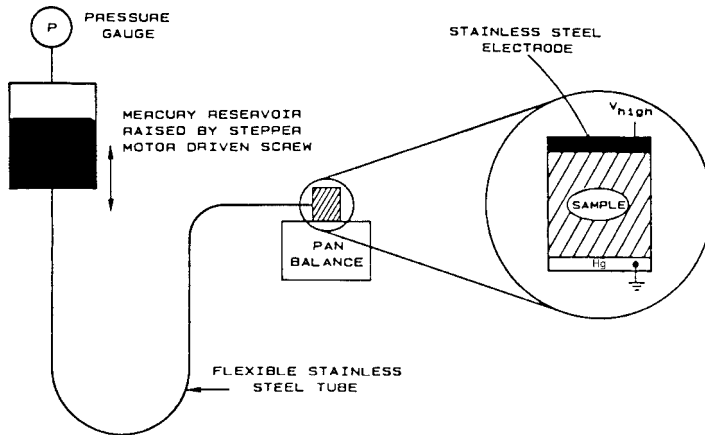


Figure 4.6 Schematic of the apparatus for mercury porosimetry (after Thompson *et al.*, 1987a).

for volume, 0.5 Pa for pressure, and $0.1 \mu\Omega$ for resistance, which result in resolutions of better than 1 part in 10^4 for all the parameters of interest. A typical experiment consists of 30 000 observations taken at intervals of 3 s.

As discussed earlier, there is a characteristic shift, or hysteresis, between the intrusion and extrusion curves. That is, the path followed by the intrusion curve is not the same as that of the extrusion curve. Similar to the drainage and imbibition experiments described above, some mercury stays in the medium even after the pressure is lowered back to atmospheric pressure. Often, hours after the pressure has been lowered back to atmospheric, some mercury continues to slowly leave the sample. In some cases, the hysteresis depends on the *history* of the system, or the way the experiment is carried out. Thus, in some cases, hysteresis can be eliminated by performing the experiment very slowly (but this is very rare), while in other cases, it *cannot* be eliminated. The latter type of hysteresis was called *permanent hysteresis* by Everett (1967).

The contact angle of mercury with a wide variety of surfaces is between 135° and 142° (the surface tension of liquid mercury at room temperature is 0.48 N/m). However, as a fluid moves or flows over a solid surface, its contact angle with the surface may vary since the fluid advances on a dry surface but recedes on a wetted surface. Thus, one may have a *contact-angle hysteresis*. Usually, the *advancing* contact angle θ_A , associated with intrusion or drainage, is larger than the *receding* contact angle θ_R that is operative during extrusion or imbibition (see Chapter 14). For mercury, a contact angle hysteresis between 10° and 20° has been reported for a variety of surfaces. Lowell and Shields (1981) showed that superposition of the intrusion and extrusion curves can be achieved if (1) the contact angle is adjusted from θ_A (or θ_i), the advancing contact angle for intrusion, to θ_R (or θ_e), the receding contact angle for extrusion, and (2) the curves are plotted as volume of the mercury versus the pore radii. For example, one may superimpose the first extrusion curve on the second intrusion curve if the contact angle for the former is also used for the latter

process. All mercury porosimetry curves have certain features in common that are as follows:

1. They all exhibit hysteresis. Thus, at a given pressure, the volume indicated on the extrusion curve is larger than the corresponding volume on the intrusion curve, whereas at a fixed volume of injected mercury, the pressure on the extrusion curve is smaller than the corresponding pressure on the intrusion curve.
2. Because some mercury stays in the porous sample at the end of the *first* intrusion–extrusion experiment, the corresponding curves do not form a closed loop.
3. If a second intrusion–extrusion experiment is carried out at the end of the first one, the corresponding curves will still exhibit hysteresis. However, the loops eventually close if the cycles continue, implying that no further entrapment of mercury in the porous medium is possible.

Why does hysteresis occur? Lowell and Shields (1982) proposed that the *pore potential* prevents extrusion of mercury from a pore until a pressure less than the nominal extrusion pressure is reached, thereby causing the hysteresis. To derive the pore potential U , one proceeds as follows. Suppose that F is the force for intrusion into a cylindrical pore (throat) of radius r (hence, $F = \pi r^2 P_i$). The pore potential is defined by

$$U = \int_0^{\ell_i} F d\ell_i - \int_0^{\ell_e} F d\ell_e, \quad (4.23)$$

where ℓ_i is the total length of mercury column in all the pores when filled, and ℓ_e is the corresponding value when mercury is extruded. Therefore, U is the difference between the work done for intruding and extruding the mercury. If $\langle r \rangle$ and $\langle \ell \rangle$ are, respectively, mean pore radius and pore length, then, given that force is the product of pressure and area of a pore, we obtain

$$U = \pi \langle r \rangle^2 (P_i \langle \ell_i \rangle^2 - P_e \langle \ell_e \rangle^2) = P_i V_i - P_e V_e, \quad (4.24)$$

implying that the pore potential is the difference between the pressure–volume work for intrusion and extrusion of mercury. Thus, intrusion of mercury into a pore with contact angle θ_i (or θ_A) results in *increased* interfacial free energy. When the pressure is lowered, mercury starts extruding from the pore at pressure P_e , reducing the interfacial area and the contact angle to θ_e (or θ_R) and, hence, reducing the interfacial free energy. This process continues until the interfacial free energy is equal to the pore potential at which extrusion ends. This also implies that the entrapped mercury is at the pore mouth, rather than at its base.

Leverett (1941) defined a reduced capillary pressure function by

$$J = \frac{P_c}{\gamma \cos \theta} \sqrt{\frac{K_e}{\phi}}, \quad (4.25)$$

where K_e is the effective permeability of the pore space. This function, named the Leverett J -function by Rose and Bruce (1949), has been found to be successful in correlating capillary pressure data originating from a specific lithologic type *within the same formation*, but it is not of general applicability. Perhaps the reason for the lack of generality is that $\sqrt{K_e/\phi}$ is not an adequate length scale for taking into account the individual differences between pore structures of various porous media. Capillary pressure curves have been reported by a large number of authors, a long list of whom is given by Dullien (1992).

Many properties of a porous medium can be obtained from its mercury porosimetry curve, namely, the volume intruded by mercury versus the capillary pressure. Extensive discussions of how such properties are estimated are given by Lowell *et al.* (2004). What follows is a summary.

4.12.1

Pore Size Distribution

While mercury porosimetry is a relatively straightforward (albeit time consuming) experiment, the interpretation of the data is not simple. The data are usually interpreted using a slight modification of Eq. (4.13) due to Washburn (1921):

$$P_c = \frac{2\sigma}{r} \cos(\theta + \varphi), \quad (4.26)$$

where P_c , the applied pressure, is just the capillary pressure between mercury and the vacuum, σ is mercury's surface tension, θ the contact angle between mercury and the pore's surface, and φ the wall inclination angle at which the pore radius is r , with $r_t \leq r \leq r_b$, with r_t and r_b being, respectively, the pore throat and the pore body radii. So long as $\theta + \varphi < \pi/2$, the interface curvature is positive. In the classical approaches of estimating the geometrical properties of a porous medium using mercury porosimetry data, no attention is paid to the pore space connectivity, and one may analyze the problem as follows.

In a mercury porosimetry experiment, the pores are filled one by one according to Eq. (4.26). Suppose that $f_V(r)$ represents the volume pore size distribution function defined as the pore volume per unit interval of pore radius. Then, when the radius of the pores into which mercury penetrates changes from r to $r - dr$ (recall that mercury begins intrusion from the largest pores and penetrates pores of increasingly smaller radii), the corresponding change in the volume is $dV = -f_V(r)dr$. Differentiating Eq. (4.26) and assuming that σ and θ are constant, one obtains, $P_c dr + r dP_c = 0$, which, when combined with the equation for dV and the result is rearranged, yields

$$f_V(r) = \frac{P_c}{r} \left(\frac{dV}{dP_c} \right) = \frac{P_c}{r} \left(\frac{\Delta V}{\Delta P_c} \right). \quad (4.27)$$

Equation (4.27) enables one to convert the cumulative V -versus- P_c curve to one for the distribution function. To use Eq. (4.27), each $\Delta V/\Delta P_c$ should be multiplied by

the capillary pressure at the upper end of the interval, with r being its corresponding pore radius according to Eq. (4.26). One may also use the mean values of P_c and r . Equation (4.27) can also be rewritten in terms of the logarithmic volume distribution function, $f_V(\ln r)$. It is then easy to see that Eq. (4.27) can be rewritten as

$$f_V(\ln r) = \frac{dV}{d \ln P_c}, \quad (4.28)$$

which reduces the wide disparities that dV/dP_c generates via Eq. (4.27).

4.12.2

Pore Length Distribution

Clearly, if $f_V(r)$ is divided by πr^2 , the cross-sectional area of a pore, the result is the distribution function $f_\ell(r)$ for the length ℓ of the pores:

$$f_\ell(r) = \frac{f_V(r)}{\pi r^2}. \quad (4.29)$$

Of course, it is implicitly assumed that the pore population in any interval is constant.

4.12.3

Pore Number Distribution

When mercury intrudes a volume ΔV of the pores in a narrow range Δr of pore radii centered about a unit radius r , the corresponding number of pores n is given by $n\ell = \Delta V/(\pi r^2)$. That is, $f_V(r) = \Delta V/\Delta r$, for two such radii, r_1 and r_2 , one must have,

$$\frac{n_1}{n_2} = \left(\frac{r_2}{r_1} \right)^2 \frac{f_V(r_1)}{f_V(r_2)}. \quad (4.30)$$

4.12.4

Pore Surface Distribution

The pore surface distribution $f_S(r)$ is defined as surface area per unit pore radius. Since $f_S(r) = (dS/dV)(dV/dr)$, and assuming a cylindrical pore for which $dS/dV = 2/r$, one obtains

$$f_S(r) = \frac{2}{r} f_V(r). \quad (4.31)$$

4.12.5

Particle Size Distribution

It has been postulated that mercury intrusion into an unconsolidated porous medium of particles can also provide information about the particles' sizes. In particular, Mayer and Stowe (1966) and Smith and Stermer (1987) analyzed mercury

porosimetry data for packings of particles. The former postulated that the breakthrough pressure P_b that mercury needs in order to intrude the voids between the particles depends on the geometry of the packing and is given by

$$P_b = \sigma \left(\frac{\ell_{lv} - \ell_{ls} \cos \theta}{A} \right), \quad (4.32)$$

where ℓ is the perimeter length of the incipient mercury “lobe”, A its cross-sectional area, and subscripts l, s, and v refer to liquid, solid, and vapor phases. For a circular opening of radius r in which the mercury perimeter is only in contact with the solid surface, $\ell_{lv} = 0$. Then,

$$P_b = -\frac{2\sigma \cos \theta}{r}. \quad (4.33)$$

Pospech and Schneider (1989) rewrote Eq. (4.33) in a more general form:

$$P_b = \frac{c\sigma}{D}, \quad (4.34)$$

where D is the diameter of the spherical particles and c is a constant. For randomly close-packed spheres (the porosity of which is about 0.377) and a mercury contact angle of 140° , $c \simeq 10.7$. In general, however, c decreases with porosity ϕ . León y León (1998) confirmed the validity of Eq. (4.34) using carbon black particles.

Smith and Stermer (1987) made a partial correction to the theory of Mayer and Stowe (1966) by postulating that the volume V_i of mercury that intrudes into a bed of polydispersed particles at any capillary pressure P_i , is the sum of volumes intruded between particles of each size D . Hence,

$$V_i = \int_{D_{\min}}^{D_{\max}} K(P_i, D) f_D(D) dD, \quad (4.35)$$

where $K(P_i, D)$ is a kernel that describes how mercury intrudes between sets of particles of a given diameter D , and $f_D(D)$ is the particle size distribution. A numerical method is then used to de-convolute integral equation (4.35) and compute the distribution function $f_D(D)$.

4.12.6

Pore Network Models

In most of the formulae derived so far, there is no provision that allows one to take into account the effect of the interconnectivity of the throats. It should, however, be clear to the reader that the sequence of the pores and throats that are filled by mercury during its intrusion into a porous medium not only depends on the pore size distribution of the pore space, but also on its topology – the way the throats are connected to one another through the pores. Thus, mercury porosimetry belongs to the class of percolation phenomena described in Chapter 3. Although the effect of pore

space connectivity on mercury porosimetry, and more generally the capillary pressure curves for any two-phase fluid system in porous media, had been appreciated for some time, it was only in 1977 that the connection between this phenomenon and percolation was recognized. Since percolation phenomena are usually studied in networks and, as described in Chapter 3, any porous medium can be mapped onto an equivalent network of pores and throats (nodes and bonds), the possibility of developing a network model of mercury porosimetry becomes obvious. Chatzis and Dullien (1977) (complete details of their work are given by Chatzis and Dullien, 1985) and Wall and Brown (1981) developed network models of mercury porosimetry, and specifically mentioned their connection with percolation. Larson (1977) and Larson and Morrow (1981) used percolation concepts to derive analytical formulae for mercury porosimetry curves (see below). Androustopoulos and Mann (1979) used 2D networks of interconnected pores to model the phenomenon, but did not mention percolation explicitly. These works recognized that a bundle of parallel capillary tubes is inadequate for modeling mercury porosimetry. For example, Larson and Morrow (1981) and Wall and Brown (1981) recognized that pores that are close to the external surface of a porous medium can be reached more easily than those deep within the medium since if a pore in the interior of the medium is to be penetrated by mercury, a connection with the external surface via the *already intruded* pore bodies and pore throats must be established. This effect was nicely demonstrated by Dullien and Dhawan (1975), who compared pore size distributions obtained by photomicrographic techniques with those inferred from mercury porosimetry data interpreted with the above assumptions. One way of decreasing the effect of interior pores is to use thin or small samples which also reduce the measurement time. However, before this is done, one must establish that the pore size distribution obtained with small or thin samples is in fact representative of the actual (much larger) porous medium. The model developed by Larson and Morrow (1981) (see below) took the effect of sample size into account.

Once one recognizes the importance of pore connectivity and accessibility, then the application of network models of porous media and the concepts of percolation theory to mercury porosimetry becomes natural. Many research groups have used such models and concepts to compute the capillary pressure curves of porous media (Larson and Morrow, 1981; Wall and Brown, 1981; Conner *et al.*, 1988; Neimark, 1984a; Chatzis and Dullien, 1985; Heiba, 1985; Lane *et al.*, 1986; Ramakrishnan and Wasan, 1984). In addition, it was recognized that, although mercury porosimetry is a percolation process, it also has certain differences with the random percolation model described in Chapter 3. Tsakiroglou and Payatakes (1990) developed a 3D network simulator in which percolation was not used explicitly, although pore interconnectivity played an essential role.

Before describing percolation and pore network models of mercury porosimetry, let us first mention a few earlier works that, although did not make explicit use of the concepts of percolation theory, were more or less percolation models since they took into account the effect of pore interconnectivity. In particular, nearly 60 years ago, Meyer (1953) had already recognized the importance of the pores' connectivity. Ksenzhek (1963) used a simple-cubic network of capillary tubes to

study the penetration of a porous medium by a non-wetting liquid. Based on a few assumptions, Ksenzhek derived formulae for various quantities of interest. In particular, a quantity that is equivalent to the percolation probability was calculated. Topp (1971) noticed that in the early theories of hysteresis phenomena, developed by Everett (1954) and others, only the pores' shapes determined the sequence by which they are filled by the intruding fluid, whereas in reality, *both* the pore geometry *and* the state of its neighboring pores are important. Topp also developed integral convolutions of the pore size distribution and a quantity that is equivalent to the accessibility function of percolation. He clearly recognized the significance of both the pore size distribution *and* the pore space accessibility.

Let us also point out an advantage of network models. During intrusion, mercury must overcome a (capillary) pressure at a *pore throat* of radius r_t given by Eq. (4.26) with $r = r_t$, whereas during extrusion, the pressure is again given by Eq. (4.26) with $r = r_b$. Therefore, a network model can provide information on the sizes (and shapes) of pore bodies and pore throats, which is not easy to obtain by other methods. We now describe a network model of mercury porosimetry in a porous medium. In the next section, we will describe how the concepts of percolation theory are utilized to model the same phenomenon. A percolation model cannot, by itself, take into account the effect of all the pore-scale phenomena, but as we show below, it provides valuable information about its behavior.

A representative example of a network model of mercury porosimetry is that of Tsakiroglou and Payatakes (1990); see also Tsakiroglou and Payatakes (1998). They represented the porous medium by a simple-cubic network of cylindrical throats and spherical pore bodies with their effective diameters selected from Gaussian distributions. The throats' length was adjusted such that the network's porosity matched that of the porous medium. As explained earlier, it is usually assumed that the mercury intrusion is controlled solely by the radii of the throats, hence implying that once a throat is filled by mercury, so also is the downstream pore. There are some experiments, however, that indicate that this may not be the case, and that mercury menisci can reach equilibrium not only at the entrance to a throat, but also at the entrance to a pore. Tsakiroglou and Payatakes allowed this possibility in their model.

Consider a meniscus that is entering a pore shown in Figure 4.7. The capillary pressure P_c is determined as a function of the position z of the contact line in the pore given by $P_c(z) = 2\sigma \cos \alpha / r(z)$, where $r(z)$ is the pore's radius at z and is given by

$$r(z) = \left[\frac{d_t^2}{4} + z \sqrt{d_p^2 - d_t^2} - z^2 \right]^{\frac{1}{2}}, \quad (4.36)$$

with $\alpha = \theta_1 - \theta$ where

$$\tan \theta_1 = \frac{dr(z)}{dz} = \frac{\sqrt{d_p^2 - d_t^2} - 2z}{2r(z)}. \quad (4.37)$$

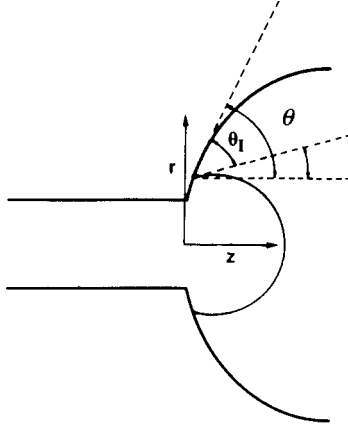


Figure 4.7 A meniscus leaving a throat and entering a pore (after Tsakiroglou and Payatakes, 1990).

Here, d_p and d_t are the diameters of the pore and the throat. If $P_c(z) \geq 4\sigma \cos \theta / d_t$, then the external pressure for moving the meniscus into the pore is larger than that for moving it into the throat.

During mercury extrusion, collars of the wetting fluid (mercury vapor and/or air) are formed in the throats that are saddle-shaped (see Figure 4.8). As the pressure is lowered during extrusion, the curvature of the collars decreases until some interfaces become unstable and rupture. This phenomenon is called *snap-off* (Mohanty *et al.*, 1980; see Chapters 14 and 15 for detailed discussions). The shape of a collar in a cylindrical throat is determined by solving the Young–Laplace equation in cylindrical coordinates,

$$r \frac{d^2 r}{dz^2} + \frac{r P_c}{\sigma} \left[1 + \left(\frac{dr}{dz} \right)^2 \right]^{\frac{3}{2}} - \left(\frac{dr}{dz} \right)^2 = 1, \quad (4.38)$$

with the boundary condition that $dr/dz = -\tan \theta_R$ at $z = 0$. Thus, varying P_c allows one to determine $r(z)$ by solving Eq. (4.38) numerically. If V_c is the volume of

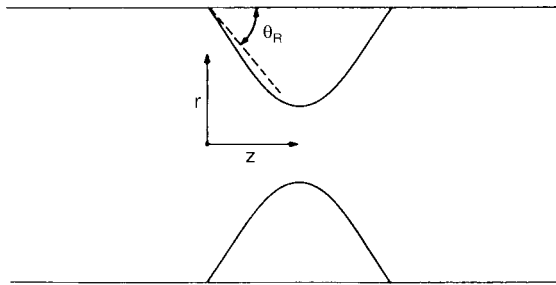


Figure 4.8 Saddle-shaped collars of the wetting fluid around the non-wetting fluid during slow imbibition (after Tsakiroglou and Payatakes, 1990).

the collar, then the collar is stable if $dV_c/dP_c \leq 0$ (Everett and Haynes, 1972). Thus, setting $dV_c/dP_c = 0$ determines the critical P_c for snap-off and the corresponding shape of the interface. Note that the length of a throat must be long enough for a collar to form.

At this point, let us mention the interesting experimental work of Wardlaw and McKellar (1981) who constructed a porous medium in which clusters of smaller elements were dispersed in a continuous network of larger elements. They found that during intrusion, the mercury invaded the largest elements, while during extrusion, it withdrew from the clusters of smaller elements. Another porous medium was then constructed in which isolated clusters of large elements were embedded in a continuous network of smaller elements. Then, although the mercury first withdrew from the smaller elements, when the pressure was reduced below the threshold for emptying of the clusters of larger elements, they had already become disconnected by snap-off of the mercury meniscus and, therefore, extensive residual mercury remained in the porous medium. These experiments indicate that the snap-off could still occur at the boundary between the two heterogeneity domains, even when the characteristic pore sizes in the two domains did not differ by more than a factor of 2–3. They also clearly indicate the significance of the spatial distribution of the heterogeneity of a porous medium to the snap-off phenomenon.

In any event, network simulation of mercury porosimetry proceeds as follows. Consider first the intrusion process and place mercury menisci at the entrance to all the throats on the network's external surface. The applied pressure is set to be small enough that no meniscus can enter a throat yet. It is then increased by a small amount, and all the menisci at the throat entrances are examined. If the applied pressure exceeds the capillary pressure of a throat, that throat is filled by mercury and the meniscus is placed at the entrance to the downstream pore. After all menisci at the pore entrances are examined and moved if necessary, the menisci at the entrances to the downstream pores are examined. If the applied pressure exceeds the capillary pressure for entering a pore, that pore is filled with mercury and new menisci are placed at the entrance to the throats that are connected to that pore but contain no mercury. If a pore has several menisci placed at its entrances, the smallest capillary pressure determines whether it is filled with mercury. The new menisci are examined to determine if any new throat can be filled with mercury. If the applied pressure is not large enough, it is increased by a small amount, all the menisci are examined again, and so on. The simulation continues until mercury-filled throat and pores form a sample-spanning cluster. Figure 4.9 shows typical configurations of a two-layer network of coordination number 5 at various stages of mercury injection and intrusion.

At the end of simulation of the intrusion process, simulation of the extrusion process begins. The applied pressure is lowered by a small amount and a search is carried out to identify (1) the pores with menisci that are connected to the external mercury sink through continuous mercury paths, and can move under the present conditions, and (2) the throats that contain mercury that must snap-off under the present conditions. For such pores, Eq. (4.38) is solved numerically to determine the shape of the interface. Snap-off can leave pockets of isolated mercury in some

throats or pores, which are then ignored for the rest of the simulation. The applied pressure is lowered again and the search continues. Clearly, simulation of the retraction process is much more difficult than the injection because one must deal with the snap-off phenomenon. Figure 4.10 presents typical configurations of the same network as in Figure 4.9, though during the retraction. Figure 4.11 shows the computed capillary pressure–saturation curves along with those for smaller networks, which resemble those obtained experimentally.

The advantage of a network model is that most of the pore-level phenomena can be taken into account and, therefore, one obtains a clear understanding of what happens during mercury porosimetry. Moreover, one can vary the pore throat and pore body size distributions, and the coordination number of the network to study their effect on mercury porosimetry. To obtain the pore throat and pore body size distributions, one can assume the two distributions to have plausible functional forms with one or a few adjustable parameters and vary them until the simulation results match the data. However, this procedure is tedious and time consuming. Tsakiroglou and Payatakes (2000) described how their network model can be combined with serial sectioning analysis of porous samples and mercury porosimetry data in order to characterize the samples.

We should point out that although a pore network model of the type described above can provide insight and information about the statistical distributions of

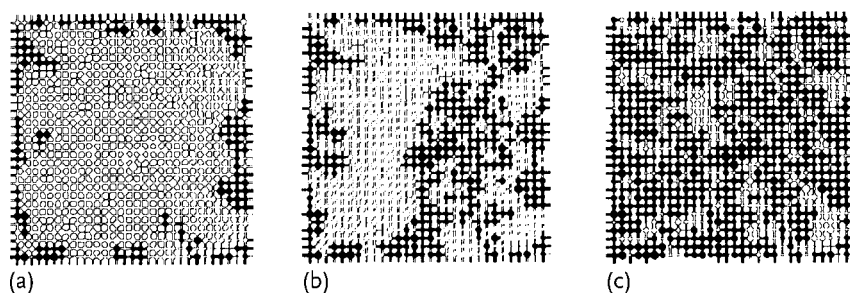


Figure 4.9 Configurations of the pore network during mercury injection. The applied pressure increases from (a) to (c) (after Tsakiroglou and Payatakes, 1990).

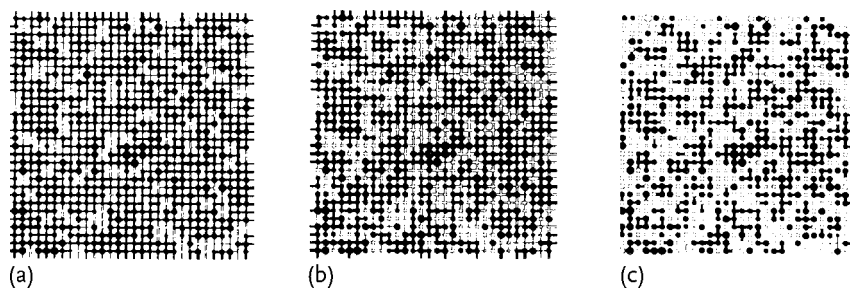


Figure 4.10 Configuration of the pore network during mercury retraction. The pressure decreases from (a) to (c) (after Tsakiroglou and Payatakes, 1990).

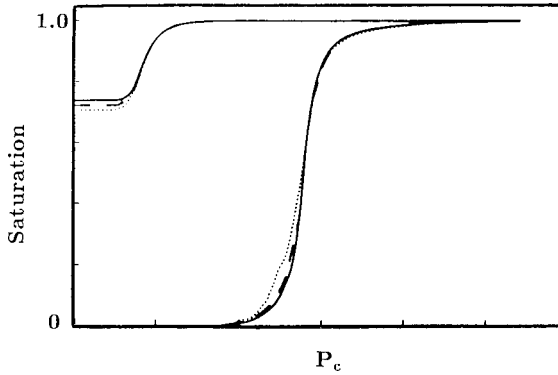


Figure 4.11 The computed capillary pressure curve. Dotted, dashed, and solid curves are, respectively, for the smallest, intermediate and largest networks (after Tsakiroglou and Payatakes, 1990).

pores' and throats' sizes, it cannot, by itself, provide any information about the *correlations* between the pores and throats. In fact, Knackstedt *et al.* (1998, 2001) showed, using a pore network model, that in order to be able to obtain *quantitative* fit of experimental data on mercury porosimetry, the effect of the correlations between the sizes of the throats must be taken into account. As the discussion in Section 4.5 indicated, the correlations are quite extended.

Let us also point out that most of the work on mercury injection into a porous medium has been done on conventional porosimetry in which the injection pressure is controlled and held constant during each step of the injection. However, rate-controlled (or volume-controlled) mercury injection experiments may provide much more information on the statistical nature of pore structures than conventional porosimetry (Yuan and Swanson, 1989; Toledo *et al.*, 1994). Fluid intrusion under conditions of constant-rate injection leads to a sequence of jumps in the capillary pressure that are associated with regions of low capillarity. While the envelope of the curve is the classic pressure-controlled curve, the invasion into regions of low capillarity adds discrete jumps onto the envelope. In the experiments of Yuan and Swanson (1989), mercury injection into a sample was done by a stepping-motor-driven positive displacement pump. This method gives a volume-controlled measurement of the capillary pressure P_c , monitored as a dependent variable. The particular sequence of alternate reversible and spontaneous changes is determined by the structure of the porous medium and the saturation history. An understanding of this relationship is essential to converting P_c fluctuations into pore-structure information. In Figure 4.12a, we show an example of a capillary pressure curve obtained for Berea sandstone under rate-controlled conditions. The detailed geometry of the jumps in the capillary pressure curve over different saturation ranges is shown in Figure 4.13b–d. Knackstedt *et al.* (1998, 2001b) showed that a proper pore network model successfully simulates the experimental results shown in Figure 4.12.

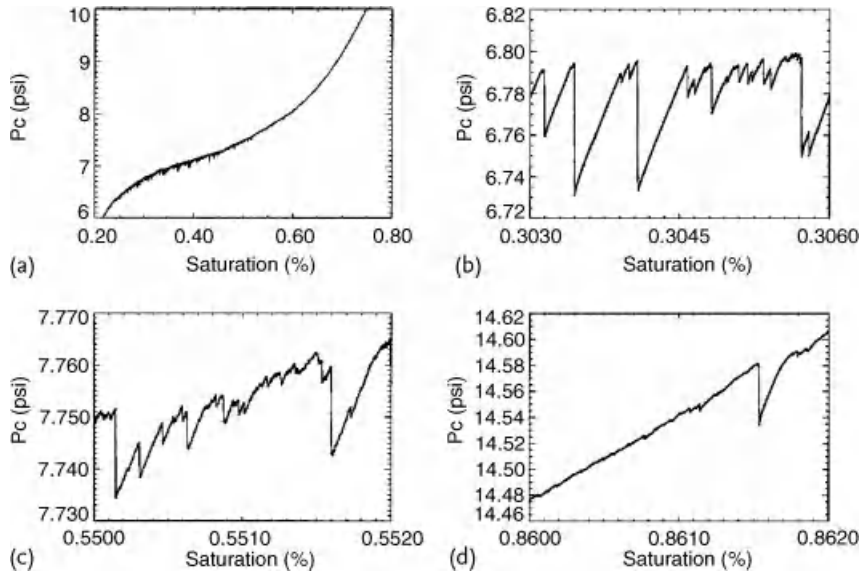


Figure 4.12 Experimental constant-volume mercury porosimetry for Berea sandstone. (a) shows the all the data and (b)–(d) show portions of them over some saturation ranges (after Knackstedt *et al.*, 2001b).

A great advantage of mercury porosimetry is that it probes a very broad range of pore sizes. At the same time, if a porous medium contains pores and throats with very broad ranges of sizes, it might become difficult to represent it by a network large enough to contain such ranges in a meaningful manner. To address this problem, multiscale and hierarchical network models were proposed. Neimark (1989), Xu *et al.* (1997), and Vocka and Dubois (2000) developed a class of such models in which the various relevant length scales were considered by examining the porous medium at various *magnifications*. As the magnification increases, information on the coarser pore system “dissipates” and, thus, one can access detailed information on the finer pores. A technique was then used to reduce the complex pore space with multiple relevant length scales to one with a single scale with the correct properties.

Rigby (2000) and Rigby *et al.* (2002) generalized the multiscale model and introduced a multiscale hierarchical model in which each level of the hierarchy consists of a square network. At the finest scale of the hierarchy that represents the pore scale, each separate site of the square network consists of cylindrical pores of equal radii, but the radii of the pores may vary from site to site. Each site at this level would contain the same pore volume. At higher levels the pore size allocated to a specific site in a square network corresponds to the average pore size for the domain of the sample represented by that particular network site. In general, the pore sizes, regardless of whether they represent the actual pores’ radii in that level or represent some average sizes at the higher level, can be distributed according to representative statistical distributions. In this way, a hierarchy of several relevant

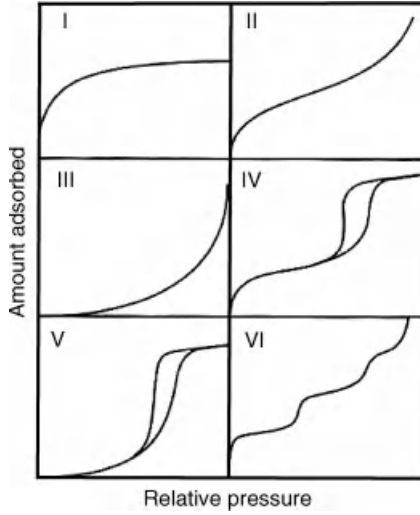


Figure 4.13 Six types of sorption isotherms (see the text).

length scales and their associated pore sizes are constructed and represented by a more general network.

Finally, we should mention the interesting work of Deng and Lake (2001) who combined a network model of pores and throats with a thermodynamic view of capillary phenomena in order to compute the capillary pressure curves. The method requires that the Helmholtz free energy of the system – the network and the two fluids that it contains – be at a minimum at all the saturation states. Among other things, the model could predict capillary pressure at arbitrary wetting states, including *negative* values of P_c .

4.12.7

Percolation Models

Let us now describe how the concepts of percolation theory may be utilized in order to develop a model for mercury porosimetry. We should first note that such concepts are applicable to describing two-phase fluid flow in porous media if the capillary pressure across a meniscus separating the two fluids (for example, mercury and the vacuum) is greater than any other pressure difference in the problem, such as, that due to buoyancy. The second condition is that frictional losses due to viscosity must be small compared with the capillary work. To quantify this condition, we define a dimensionless group, called the *capillary number* Ca , by

$$Ca = \frac{\mu v}{\sigma}, \quad (4.39)$$

where v is the average fluid velocity and μ the average viscosity. Then, one must have $Ca \ll 1$ in order to fulfill the second condition. The porous medium is again

represented by a network of interconnected cylindrical throats connected to one another at the pores (sites). The effective radii of the throats are distributed according to statistical distribution $f(r)$. We describe two percolation models that are, however, closely related.

The first model that we describe was developed by Larson and Morrow (1981). In their model, one writes the capillary pressure as $P_c = \sigma C$, where C represents the curvature of a meniscus (as $C \propto 1/r$). Consider, first, the injection process. The network is exposed to mercury under an applied pressure that, given that the surface tension σ is constant, is equivalent to being exposed to some meniscus curvature C . As C (P_c) increases, it exceeds the entry curvature C_i (entry pressure P_i) for a larger and larger fraction of the throat. In fact, C exceeds C_i for a fraction $Y_i(C)$ of the void volume given by

$$Y_i(C) = \int_0^C \gamma_i(C_i) dC_i . \quad (4.40)$$

Here, $\gamma_i(C_i)$ is the *pore entry-curvature* distribution given by

$$\gamma_i(C_i) = \int_0^\infty \gamma(C_i, C_w) dC_w , \quad (4.41)$$

where $\gamma(C_i, C_w)$ is the *joint* probability distribution for throat entry and withdrawal curvatures, C_i and C_w , with the properties that $\gamma(C_i, C_w) \geq 0$ for $C_i \geq 0$ and $C_w \geq 0$ and $\gamma(C_i, C_w) = 0$ for $C_i < C_w$. Moreover, since $\gamma(C_i, C_w)$ is a probability distribution, it must be normalized so that

$$\int_0^\infty \int_0^{C_i} \gamma(C_i, C_w) dC_i dC_w = 1 . \quad (4.42)$$

Given the physical meaning of $\gamma_i(C_i)$, $Y_i(C)$ is the fraction of the throats with entry curvatures less than C . However, not all the throats with an entry curvature C are *accessible* from the external surface of the network where accessibility is defined in the percolation sense. Thus, Larson and Morrow (1981) assumed that the saturation of mercury (the non-wetting fluid) is given by

$$S_{nw} = X^A [Y_i(C)] . \quad (4.43)$$

As explained below, Eq. (4.43) is only a rough estimate of S_{nw} because one must take into account the effect of the size distribution of the throats. Therefore, for any given capillary pressure, $P_c = \sigma C$, the fraction $Y_i(C)$ is calculated based on which the saturation S_{nw} is determined.

Next, consider the retraction (extrusion) process. As the applied pressure at the end of the injection process decreases, so also does the curvature C that can *potentially* eject mercury (the non-wetting fluid) from those throats for which the withdrawal curvature is between C_w and $C_w - dC_w$, and the entry curvature is C_i or

less. The fraction of such throats is

$$\gamma_r(C_i, C) = \int_0^{C_i} \gamma(C'_i, C) dC'_i . \quad (4.44)$$

However, not all such throats actually expel mercury because some of them were not invaded during the injection process in the first place as they were not accessible, while some others, although containing mercury, cannot expel it because they are not connected to the external surface of the network via a path of mercury-filled throats. Thus, during retraction, the fraction of the throats with injection curvature less than or equal to C_i and withdrawal curvature less than or equal to C_w is given by

$$Y_r(C_i, C) = \int_0^C \gamma_r(C_i, C_w) dC_w . \quad (4.45)$$

Only a portion $X^A(Y_r)$ of such throats still contain mercury and are also accessible, implying that their fraction is $X^A(Y_r)/Y_r$. Thus, as the curvature C_w is reduced by an amount dC_w (as the applied pressure is reduced), the saturation of mercury also decreases by dS_{nw} given by

$$-dS_{nw} = \frac{X^A[Y_r(C_i, C_w)]}{Y_r(C_i, C_w)} \gamma_r(C_i, C_w) dC_w , \quad (4.46)$$

which, after integration, yields

$$S_{nw} = S_0 - \int_C^{C_i} \frac{X^A[Y_r(C_i, C_w)]}{Y_r(C_i, C_w)} \gamma_r(C_i, C_w) dC_w , \quad (4.47)$$

where S_0 is the initial saturation at which the retraction process started. Therefore, given a capillary pressure P_c , one determines the corresponding saturation S_{nw} . Clearly, when the applied pressure P_c is zero, so also is the corresponding curvature which is the point at which the saturation of mercury (the non-wetting fluid) is at its residual value, S_{rnw} , determined from Eq. (4.47) by setting $C = 0$. This completes computation of the retraction curve.

At the end of the retraction (extrusion) process, we may consider a *second* injection process for which it is not difficult to see that one must have

$$\gamma_i(C_i, C) = \int_0^C \frac{X^A[Y_r(C_i, C_w)]}{Y_r(C_i, C_w)} \gamma(C, C_w) dC_w , \quad (4.48)$$

using the fact that, for $C < C_w$, one has $\gamma(C, C_w) = 0$. The corresponding saturation is given by

$$S_{nw} = S_{rnw} + \int_0^C \gamma_i(C_i, C') dC' . \quad (4.49)$$

Clearly, one may also consider a secondary retraction process.

For a given curvature distribution, one has a corresponding throat size distribution as $r \propto 1/C$. Therefore, given a porous medium and its capillary pressure curves, one may assume a functional form for the throat size distribution $f(r)$ with a few adjustable parameters which are estimated such that the predicted capillary pressure curves agree with the data. The advantage of the model just described is that if the accessibility function X^A (which depends on the connectivity of the pore network) is available, then computing the capillary pressure curves will be straightforward; there would be no need for detailed pore network simulations of the type described in the last section. For 3D networks, however, no closed-form formula is known for X^A ; only its numerical values have been computed. Thus, the integrals in Eqs. (4.46)–(4.48) must be numerically evaluated, a task that is still simpler than the computations associated with the pore network simulation described in the last section.

To simplify their computations, Larson and Morrow (1981) assumed that the pore space can be represented by a Bethe lattice of coordination number 4 (see Chapter 3) that, although not realistic, has the advantage that its $X^A(p)$ is known in closed form. For a Bethe lattice of coordination number Z and $p \geq p_c$, one has (Larson and Davis, 1982),

$$X^A(p) = p [1 - R(p)^{2Z-2}] , \quad (4.50)$$

where $R(p)$ is the root of the equation, $p \sum_{j=2}^{Z-1} R^{Z-j} + p - 1 = 0$. Furthermore, Larson and Morrow (1981) assumed that

$$\gamma(C_i, C_w) = 6g(C_i)g(C_w) \int_{C_w}^{C_i} g(C) dC , \quad C_i \geq C_w \quad (4.51)$$

which possesses all the properties that were described above, and that, $g(x) = 2x \exp(-x^2)$ with $x = \log C$, which results in a broad distribution of the curvatures and, therefore, a broad $f(r)$. Figure 4.14 compares the predictions of the model of Larson and Morrow with mercury porosimetry data for a Becher dolomite with porosity, $\phi = 0.174$. The qualitative agreement between the predictions and the data is striking. Also shown is the effect of sample size (thickness) on the capillary pressure curves. The most important reason for the success of the model is the fact that the physical concept of accessibility that determines which pores can be invaded by mercury, and from which pores it can be withdrawn, has been explicitly utilized. Clearly, any pore network simulator of the type described in the last section also incorporates the concept of accessibility which is fundamental to modeling of multiphase flow phenomena in porous media.

The second percolation model, due to Heiba *et al.* (1982, 1992), represents a refinement of the model of Larson and Morrow (1981). The basis for Heiba *et al.*'s model is that during injection and retraction the spatial distributions of the pores accessible to and occupied by mercury, which we refer to as the *sub-distributions*,

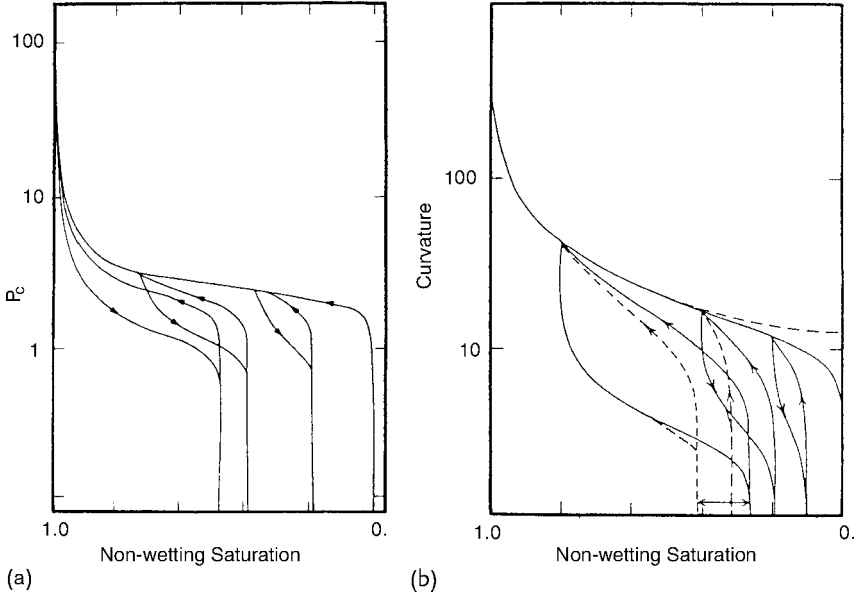


Figure 4.14 Comparison of capillary pressure data (a) with the predictions of the percolation model (b). Dashed curves are for a pore network that is 10 pore thick, while the predicted solid curves are for an infinitely large network (after Larson and Morrow, 1981).

are not identical. Consequently, the throat size distribution of the subset of pore space occupied by mercury differs from the *overall* throat size distribution. Heiba *et al.* derived analytical formulae for such sub-distributions. For example, during injection the fraction of throats that are *allowed* to mercury (the throats with the right P_c that can potentially be filled by mercury) is

$$Y_i(r_{\min}) = \int_{r_{\min}}^{\infty} f(r) dr, \quad (4.52)$$

where $r_{\min}$ is the minimum throat radius into which the mercury can penetrate. The fraction of the throats that are accessible to, and thus occupied by, the mercury is $X^A(Y_i)$. Therefore, during injection, the distribution $f_i(r)$ of the throat radii that are occupied by mercury is (Heiba *et al.*, 1982, 1992)

$$f_i(r) = \frac{f(r)}{Y_i(r_{\min})}, \quad r \geq r_{\min}, \quad (4.53)$$

and, clearly, $f_i(r) = 0$ for $r < r_{\min}$. The basis for Eqs. (4.52) and (4.53) is that during injection, the *largest* throats (i.e., those with the *smallest* entry curvature or capillary pressure) are occupied (which can be understood by examining Eq. (4.5)).

During injection, the mercury (non-wetting fluid) saturation is given by

$$S_{nw} = \frac{X^A[Y_i(r)]r_{min} \int_{r_{min}}^{\infty} f(r) V_p(r) dr}{Y_i(r_{min}) \int_0^{\infty} f(r) V_p(r) dr}, \quad (4.54)$$

where $V_p(r)$ is the volume of the pore throat of radius r . Equation (4.54) should be compared with Eq. (4.43).

As the pressure decreases during retraction, mercury is first expelled from the *smallest* throats. (In reality, mercury is expelled from the smallest pores, but the model ignored the pores.) The *allowed* fraction of such throats is (Heiba *et al.*, 1982, 1992)

$$Y_r = \int_0^{r_0} f(r) dr + \left[1 - \frac{X^A(Y_{i,t})}{Y_{i,t}} \right] \int_{r_0}^{\infty} f(r) dr, \quad (4.55)$$

where r_0 is the throat's radius at a given capillary pressure P_c , such that mercury is expelled from all the throats for which $r \leq r_0$, $Y_{i,t} = Y_i(r_{min,t})$, and $r_{min,t}$ is the throat radius at the end of injection. The first term of the right side of Eq. (4.55) is simply the fraction of the throats from which mercury is expelled if at the end of injection there were no throats that were not inaccessible to it. However, at the end of injection, a fraction $1 - X^A(Y_{i,t})/Y_{i,t}$ of the throats could not be reached by mercury and, consequently, the second term of the right side of Eq. (4.55) is the fraction of throats that were not invaded by mercury at the end of injection. Hence, the size distribution of the throats from which mercury is expelled is given by (Heiba *et al.*, 1982, 1992)

$$f_r(r) = \begin{cases} \frac{f(r)}{Y_r} \left[1 - \frac{X^A(Y_{i,t})}{Y_{i,t}} \right], & r > r_0 \\ \frac{f(r)}{Y_r} \left\{ 1 - \frac{X^A(Y_{i,t})}{Y_{i,t}} \left[1 - \frac{X^A(Y_r)}{Y_r} \right] \right\}, & r_{min,t} < r < r_0. \end{cases} \quad (4.56)$$

Clearly, $f_r(r) = f(r)/Y_r$ for $r < r_{min,t}$. Therefore, the model of Heiba *et al.* classifies the throats more carefully than that of Larson and Morrow (1981) and, moreover, calculates the saturation correctly as it takes into account the effect of the size distribution of the throats and the dependence of their volume on their effective radii.

The procedure for using the above model to extract the throat size distribution of the pore space is as follows. First, a functional form for $V_p(r)$ and, hence, a throat shape must be assumed. Next, one must have, or compute, the accessibility function of the pore space for which either the average coordination number $\langle Z \rangle$ of the pore space must be known from measurements, or it must be treated as an adjustable parameter in order to fit the percolation model to the data. (Later

in this chapter, we will describe how a combination of the concepts of percolation and sorption isotherms in porous media can be used for estimating $\langle Z \rangle$.) Observe that S_{nw} can be measured, and that P_c is simply the applied pressure set during the experiment and, thus, r_{min} can be determined. Therefore, by assuming an $f(r)$ as an initial guess, Eq. (4.54) is iterated many times until a satisfactory fit of the experimental data to the predictions is obtained. As before, a particular form of $f(r)$ with a few adjustable parameters is assumed.

At this point, it is important to clearly state and understand all the assumptions that were made in order to develop the above percolation models of mercury porosimetry:

1. The pore space is infinitely large.
2. The entire process is described by a random bond percolation.
3. Entrapment of mercury in isolated clusters is ignored.

The first assumption is essential if one is to use the results for percolation that are typically defined for infinitely large networks. However, for a given network, we may calculate X^A as a function of its linear size L . The main effect of the size or thickness is that the pore space of thinner samples are better accessed, and also reduce the sharpness of the injection-curve knee. Experiments show that injection curves for (unconsolidated) packings depend rather strongly on sample thickness for packings of up to about 10 particle diameters or about 30 throat diameters. For thicker samples, the thickness-dependence of the curves is relatively weak. In fact, if the thickness exceeds 20 particle diameters, no appreciable sample size effect can be detected.

The second assumption is not, strictly speaking, correct. As discussed in the last section, there is some evidence that once mercury fills a throat, the corresponding meniscus does not necessarily enter the downstream pore and, moreover, pores largely control the retraction process and, therefore, retraction is a site percolation rather than bond percolation problem. Therefore, a correct percolation model of mercury porosimetry should involve a mixture of bond and site percolation, and the size distributions of *both* pores and throats, whereas the above formulae were derived assuming a size distribution for the throats, ignoring the pores. The assumption that the entire process is a classical *random* percolation is also not, strictly speaking, correct since in practice, the pore space is invaded by mercury from its external surface and, therefore, the phenomenon is an *invasion percolation* process that will be described in Chapter 15. However, as discussed there, the error caused by the assumption of mercury porosimetry being a random percolation process is small.

Finally, although the third assumption is not completely correct, the resulting error is not large because, although one must consider a percolation problem in which trapping of clusters of one fluid is allowed if they are completely surrounded by clusters of another fluid – a problem that was first studied by Sahimi (1985) and Sahimi and Tsotsis (1985) in the context of catalytic pore plugging – computer simulations (Dias and Wilkinson, 1986) indicated that in 3D networks, the effect

of trapping is small enough to be neglected. Despite such shortcomings and criticism, the percolation models have been relatively successful in describing mercury porosimetry.

4.13

Sorption in Porous Media

Another important and popular method of determining the pore size distribution and surface area of pores is based on sorption isotherms of a gas, a method first suggested by Barrett *et al.* (1951). Normally, nitrogen is used in such experiments although one can, in principle, use other gases such as CO₂. Extensive discussions of gas sorption in porous media are given by Dabrowski (2001). Gas sorption allows one to characterize the pore size distribution over a wide range, from about 0.35 nm to over 100 μm . In fact, if we combine gas adsorption with mercury porosimetry, we can probe pores as large as 400 μm , hence spanning the entire nano-, meso-, and macropore regions. To begin describing the method and discussing its various aspects, let us set our terminology: The solid surface on which a gas is adsorbed is called the *adsorbent*; the gas to be adsorbed is referred to as the *adsorptive*, and when the gas is in the adsorbed state, we call it the *adsorbate*. The amount of gas that can be adsorbed on a solid surface depends on the temperature and pressure of the system as well as the interaction energy E between the adsorbates and the adsorbent. Adsorption isotherm is a plot of the amount adsorbed versus the (relative) pressure. Generally speaking, we divide adsorption processes into two distinct groups:

1. In the first group is what we call *physical* or *reversible* adsorption, also referred to as *physisorption*, which happens when an adsorbable gas is brought into contact with a solid surface. It (1) is, in most cases, accompanied by low heat of adsorption; (2) is fully reversible; (3) reaches equilibrium rather quickly since it requires no activation energy; (4) can lead to multilayer adsorption, and (5) can fill the pores completely.
2. *Chemisorption*, or *irreversible* adsorption processes, are in the second group. In this case, there are large interaction potentials between the adsorbates and the solid surface, hence leading to large heats of adsorption as well. The high heat of adsorption approaches the energies for the formation of chemical bonds and, therefore, chemisorption involves formation of such bonds between the adsorbates and adsorbents, which usually happens at temperatures *above* the critical temperature of the adsorbate. Hence, chemisorption usually leads, by necessity, to the formation of only a monolayer of adsorbate on the solid surface. Formation of chemical bonds also restricts the mobility of adsorbates on the adsorbents, whereas in physisorption, the adsorbates can move more freely on the surface.

4.13.1

Classifying Adsorption Isotherms and Hysteresis Loops

According to the International Union of Pure and Applied Chemistry (IUPAC), all sorption isotherms fall into one of the six classes shown in Figure 4.13. Two of them represent sorption isotherms with hysteresis, while the other four represent completely reversible adsorption. In Type I isotherm, which is obtained when adsorption is limited to *at most* a few molecular layers, the adsorbed amount reaches a limiting value when $P/P_0 \rightarrow 1$, where P and P_0 are, respectively, the condensation and saturation pressures. True chemisorption which can only form a monolayer, and adsorption in nanoporous materials, exhibit this type of isotherms. Type II isotherms are exhibited by porous materials that contain macropores so that the adsorbed amount can continue to increase. The inflection point of the curve indicates the point at which monolayer adsorption has ended and multilayer sorption has begun.

Type III isotherms are rare (examples include water adsorption on the clean basal plane of graphite and nitrogen adsorption on polyethylene). They indicate that attractive adsorbate–adsorbent interactions are weak, while molecular interactions between the adsorbates themselves are strong. Mesoporous materials typically exhibit Type IV isotherms, with their inflection point again signifying the end of monolayer, and the beginning of multilayer, formation. Hysteresis loops are formed as described below. Type V isotherms are similar to Type IV, except that their initial part is similar to Type III.

Type VI isotherms are unusual in that they exhibit a stepwise structure associated with uniform, non-porous surface. The sharpness of the steps depends on the homogeneity of the surface, the adsorbate type, and the temperature. An exam-

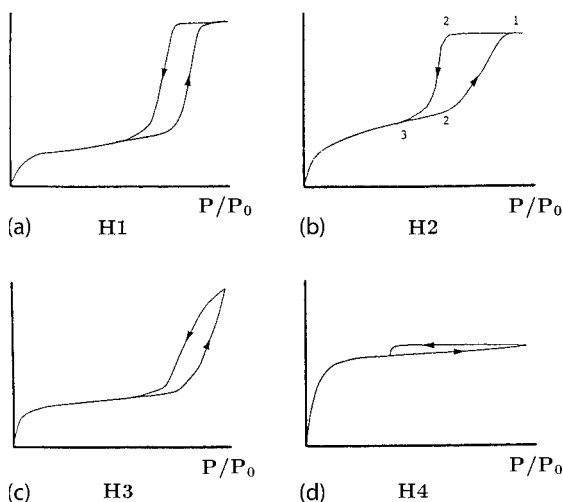


Figure 4.15 Hysteresis loops in sorption experiments.

ple is adsorption of argon and krypton on graphite at liquid nitrogen temperature (Greenhalgh, 1967).

The IUPAC has also classified the various hysteresis loops that can be observed experimentally as types H1, H2, H3 and H4, which are shown in Figure 4.15. According to Sing *et al.* (1985), at least for types H1, H2, and H3, the connectivity of the pore space plays an important role. We will come back to this point shortly.

4.13.2

Mechanisms of Adsorption

The mechanisms by which molecules are adsorbed depend on whether adsorption takes place on an open surface or in the pores of a porous medium. In addition, adsorption in the pores depends on the sizes of the pores and the adsorbates, the interactions between the adsorbates themselves, and between them and the walls of the pores, and the temperature and pressure of the system. In what follows, we describe mechanisms of adsorption in a porous material.

4.13.2.1 Adsorption in Micropores

During adsorption, mesopores are filled by the adsorbates through pore condensation that represents a *first-order* gas–liquid phase transitions. Micropores, on the other hand, are usually filled up by a continuous or *second-order* process. In this case, strong interactions of the gas phase with the pore’s walls play a fundamental role in the adsorption process. Other important factors include the molecular size and shape of the molecules and those of the pore. If a porous material is made solely of micropores, then it would exhibit Type I isotherms. However, most microporous materials contain a range of mesopores as well, in which case they exhibit isotherms with features from both Types I and IV.

Due to the small size of a micropore and its proximity to the molecular size of adsorbates, atomistic simulations, such as molecular dynamics and Monte Carlo simulations, and the density-functional theories represent the most realistic and accurate methods for studying adsorption in micropores. The description of such methods is beyond the scope of this book. The reader is referred to comprehensive reviews by Gelb *et al.* (1999) and Sahimi and Tsotsis (2006).

4.13.2.2 Adsorption in Mesopores: The Kelvin Equation

Adsorption in mesopores not only depends on the interaction between the adsorbates and the pores’ walls, but also between the adsorbates themselves. Thus, as mentioned earlier, one has condensation – a first-order phase transition by which a gas makes a transition to a liquid-like state at a pressure less than the saturation pressure P_0 of the bulk liquid. Due to the size of the pores and the condensation phenomenon, one usually has multilayer adsorption in mesopores, with the adsorption isotherms being of Types IV and V. It should be clear to the reader that the condensate must completely wet the pores’ surface in order to form a layer.

Consider, for example, Type IV isotherms for adsorption/desorption shown in Figure 4.13. When the relative pressure P/P_0 is low, adsorption in a mesopore is similar to one on a flat surface. The knee of the curve in the figure denotes completion of a monolayer of the condensate on the pore's surface. Right before the adsorption isotherm takes off sharply, multilayers have formed and attained a critical thickness. Then, pore condensation – a first-order discontinuous transition – occurs which causes a large jump in the isotherm. When the pore is completely filled with the liquid-like adsorbate, the plateau region is reached. This region is then separated from the gas phase by a meniscus. Then, on the desorption part of the diagram, evaporation of the condensate in the pores takes place and the meniscus recedes. Once again, a sharp jump reduces the adsorbed amount. The hysteresis ends when the pressure is equal to the pressure at which the multilayers had formed during adsorption and is in equilibrium with the bulk gas pressure. It is generally believed that two mechanisms contribute to the hysteresis:

1. The first mechanism has its roots in thermodynamics. A metastable phase may exist beyond the coexistence pressure during adsorption and/or desorption. This means that during adsorption, a vapor phase may exist at pressures above condensation pressure P , or during desorption a liquid phase may exist below P . Thus, this is a pore-scale mechanism and has nothing to do with the connectivity of the pore space, and gives rise to H1-type hysteresis.
2. In the second mechanism, the geometry and interconnectivity of a pore do matter. A pore with an effective radius r is allowed to desorb (to contain vapor) if not only is its radius r large enough that at a given pressure the liquid phase in it is metastable with respect to the vapor phase, but also has access to either the bulk vapor in primary desorption, or the isolated vapor pockets in secondary desorption that occurs after the secondary adsorption. The second effect is, therefore, a network-scale (pore space-scale) mechanism. It has been argued by Ball and Evans (1989) that unless the pore size distribution of the pore space is very narrow, the network mechanism is more important in the formation of the hysteresis loops. Typical adsorption–desorption isotherms of this type are shown in Figure 4.16 and give rise to type H2 hysteresis.

Consider an adsorbed, liquid-like film of chemical potential μ_a in equilibrium with the bulk gas phase at chemical potential μ_0 . Then,

$$\Delta\mu = \mu_a - \mu_0 = -RT \ln \left(\frac{P}{P_0} \right). \quad (4.57)$$

For a liquid-gas interface to coexist in a pore, one must have $\Delta\mu = P_c/\Delta\rho$, where P_c is the capillary pressure at the gas–liquid interface and $\Delta\rho = \rho_l - \rho_g$, with ρ_l being the liquid density at bulk condition and ρ_g the gas density. Therefore, using Eq. (4.14), we obtain

$$\ln \left(\frac{P}{P_0} \right) = - \left(\frac{1}{r_1} + \frac{1}{r_2} \right) \frac{\sigma \cos \theta}{\Delta\rho}. \quad (4.58)$$

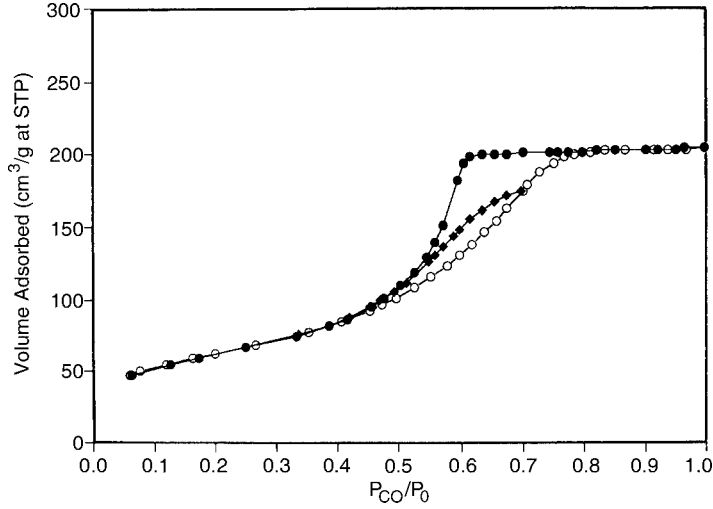


Figure 4.16 Nitrogen sorption isotherms for a porous alumina sample: Adsorption (open circles), desorption (solid curves), and secondary desorption (solid diamonds) (after Liu *et al.*, 1993; courtesy of Professor N.A. Seaton).

For a cylindrical pore of radius r , $r_1 = r_2 = r$, and for complete wetting, $\theta = 0$. The density of the liquid is typically much larger than that of the gas and, therefore, $\Delta\rho \simeq \rho_l$. Then, if $V_l = 1/\rho_l$ is the molar volume of the liquid, we obtain

$$\frac{P}{P_0} = \exp \left[-\frac{2\sigma V_l}{rRT} \right], \quad (4.59)$$

which is the well-known Kelvin⁴⁾ equation.

However, Eq. (4.59) does not take into account the existence in the pore of a multilayer film prior to condensation. If the film's thickness is ℓ_f , then the Kelvin equation is modified to

$$\frac{P}{P_0} = \exp \left[-\frac{2\sigma V_l}{RT(r - \ell_f)} \right], \quad (4.60)$$

which is sometimes referred to as the modified Kelvin equation. According to Eqs. (4.59) and (4.60), for every relative pressure P/P_0 , the adsorption process is uniquely characterized by an effective pore radius r_a . Hence, adsorption (desorption) corresponds to an increase (decrease) in r_a . During adsorption, all the pores are equally accessible, the adsorbate condenses in all the pores of size $r > r_a$, and a liquid-like fluid fills the pores. For $r < r_a$, the pores fill rapidly and continuously. Thus, during *primary* adsorption, the connectivity of the pores plays no role. All that matters is the effective size of the pores.

4) William Thomson, 1st Baron Kelvin, or Lord Kelvin (1824–1907) was an Irish mathematical physicist and engineer who made many important contributions to electricity and development of the first

and second law of thermodynamics. His name is associated with many phenomena, including Joule–Thomson effect, Kelvin waves, Kelvin–Helmholtz instability and mechanism, and Kelvin equation.

4.13.3

Adsorption Isotherms

We now describe and discuss several well-known approaches for the modeling of adsorption isotherms.

4.13.3.1 The Langmuir Isotherm

As described earlier, Type I isotherms usually arise when adsorption is limited to a monolayer on the surface. Based on a monolayer assumption, Langmuir (1918)⁵ was able to derive an analytical expression for Type I isotherms. We consider a smooth surface and assume that the system is at low pressure. Suppose that N is the number of molecules hitting a unit area of the surface per unit time, and θ_v is the vacant fraction of the surface. Then, $dN/dt = k P \theta_v$ where k is a constant that, according to the kinetic theory of gases, is given by, $k = \mathcal{N} / \sqrt{2\pi M R T}$, with M being the molecular weight of the gas molecules, and $\mathcal{N}$ the Avogadro's number. The number of adsorbed molecules (per unit area) is

$$N_a = k P C_1 \theta_v, \quad (4.61)$$

where C_1 is the condensation coefficient that is interpreted as the probability that a molecule is adsorbed on the surface. We must also consider the number of molecules N_d that are desorbed, which is given by

$$N_d = N_t \theta_a \nu \exp\left(-\frac{E}{RT}\right). \quad (4.62)$$

Here, N_t is the number of adsorbed molecules in a completed monolayer of unit area, $\theta_a = 1 - \theta_v$ is the fraction of the surface covered with adsorbed molecules, ν the vibrational frequency of the adsorbate normal to the surface, and E is the adsorption energy. The terms $\exp(-E/RT)$ is the probability that a molecule to be desorbed overcomes the attractive potential of the surface. At equilibrium, $N_d = N_a$. Therefore, if $K = k C_1 \exp(E/RT)/(N_t \nu)$, we obtain

$$\theta_a = \frac{K P}{1 + K P}. \quad (4.63)$$

For monolayer adsorption, $\theta_a = N/N_t = W/W_t$, where N is the number of molecules in the incomplete monolayer, and W is their corresponding weight. Hence, substituting for θ_a in Eq. (4.63) and rearranging the result yield

$$\frac{P}{W} = \frac{1}{K W_t} + \frac{P}{W_t}, \quad (4.64)$$

5) *Irving Langmuir* (1881–1957) was an American scientist who proposed a theory of atomic structure and made many important contributions to surface chemistry for

which he received the 1932 Nobel Prize in Chemistry. The journal *Langmuir* was named after him.

which implies that a plot of P/W versus P must be a straight line of slope $1/W_t$ and intercept $1/K W_t$. The surface area of the sample covered is then given by

$$S = N_t A_c = \frac{W_t \mathcal{N} A_c}{M}, \quad (4.65)$$

where A_c is the cross-sectional area of the adsorbates. Making an independent estimate of A_c to be used in Eq. (4.65) is, by itself, an important problem that has been studied by many researchers. We mention one reasonable approximation due to Emmett and Brunauer (1937), which is given by

$$A_c = c \left(\frac{V_1}{\mathcal{N}} \right)^{\frac{2}{3}}, \quad (4.66)$$

where $c = 1.091 \times 10^{16}$, and A_c is in $\text{\AA}^2$ per molecule.

4.13.3.2 The Brunauer–Emmett–Teller (BET) Isotherm

Brunauer, Emmett, and Teller (1938) analyzed multilayer adsorption by assuming that the uppermost molecules in adsorbed layers are in dynamic equilibrium with the vapor. Because the equilibrium is dynamic, the location of the surface sites covered by one or more layers may vary, but the number of molecules in each layer will be constant. For the first layer at equilibrium, one has $N_t \theta_1 \nu_1 \exp(-E_1/RT) = k P \theta_1 C_1$, where the notation is as before except that we have used subscript 1 to denote the values of the parameters for the first layer. Similarly, for the second layer, we have $N_t \theta_2 \nu_2 \exp(-E_2/RT) = k P \theta_2 C_2$, and, in general, $N_t \theta_n \nu_n \exp(-E_n/RT) = k P \theta_n C_n$ for the n th layer. It is assumed in the BET theory that the parameters ν , E , and C remain constant for the second and higher layers, which is justifiable if these layers are all equivalent to the liquid state. Thus, if H is the heat of liquefaction, then in the equations for the second and higher layers, the energy E_i is replaced by H so that $N_t \theta_n \nu \exp(-H/RT) = k P \theta_n C$ for $n = 2, 3, \dots$. Solving, as before, for θ_n , recognizing that the total number N of adsorbed molecules is given by $N = \sum_{i=1}^{\infty} i \theta_i$, and setting $C = (C_1/C_2)(\nu_2/\nu_1) \exp[(E_1 - H)/RT]$, we finally obtain an expression for $W/W_t = N/N_t$ that after rearranging is given by

$$\frac{1}{W \left(\frac{P}{P_0} - 1 \right)} = \frac{1}{W_t C} + \frac{C - 1}{W_t C} \left(\frac{P}{P_0} \right). \quad (4.67)$$

In a pore, only a limited number of adsorbed layers can form. If n is the number of such layers in a pore, then

$$\frac{W}{W_t} = \frac{C}{\frac{P}{P_0} - 1} \frac{1 - (n + 1) \left(\frac{P}{P_0} \right)^n + n \left(\frac{P}{P_0} \right)^{n+1}}{1 + (C - 1) \frac{P}{P_0} - C \left(\frac{P}{P_0} \right)^{n+1}}. \quad (4.68)$$

From the slope and intercept of the BET isotherm, the surface area S is estimated using Eqs. (4.65) and (4.66).

Despite some criticism of the BET theory (for example, the suspect assumption that the heat of adsorption of the second and higher layers is equal to the heat of liquefaction) due to its simplicity and the ability to accommodate each of the isotherm types, the BET isotherm is almost universally used. When $n = 1$, one recovers the Langmuir isotherm. Plots of W/W_t versus P/P_0 conform to Type II or III isotherms for $C > 2$ and $C < 2$, respectively. Types IV and V isotherms are simple modifications of Types II and III isotherms due to the presence of pores. Moreover, in the range, $0.05 \leq P/P_0 \leq 0.3$, that is, near a point where a monolayer is close to completion, the BET isotherm and experimental data agree very well, which means that the surface area is estimated rather accurately.

4.13.3.3 The Frenkel–Halsey–Hill Isotherm

Although the BET isotherm is supposed to be valid for multilayer adsorption, it provides a satisfactory description of the phenomenon for only the first two or three layers. If the adsorbed layer is thick, one may consider it as a slab of liquid that has the same properties as the bulk liquid would have at the same temperature (assumed to be below the critical temperature). This was analyzed by Frenkel (1946), Halsey (1948), and Hill, T.L. (1952), commonly referred to as the FHH isotherm, and is given by

$$\ln\left(\frac{P}{P_0}\right) = \alpha \left(\frac{V_a}{A}\right)^{-m}, \quad (4.69)$$

where α is an empirical parameter, A is the total surface area, and V_a is the volume of the adsorbed phase. Thus, a plot of $\ln \ln(P/P_0)$ versus $\ln(V_a/a)$ should be a straight line. Molecular theories predict $m = 3$, although experiments usually yield $m \simeq 2.5\text{--}2.7$. The FHH isotherm is applicable only when the pressure is relatively high so that the assumption of representing the adsorbate as a slab of liquid can be justified.

4.13.4

Distributions of Pore Size, Surface, and Volume

Many methods have been proposed for using sorption isotherms in a porous medium to extract the pore volume and surface, and the pore size distribution. A review of such methods is given by Lowell *et al.* (2004). Notable among them is the Dubinin–Radushkevich method (Dubinin and Radushkevich, 1947) according to which the volume V_m of micropores is estimated from the following equation,

$$\log W = \log(V_m \rho) - c_1 \left[\log\left(\frac{P_0}{P}\right) \right]^2, \quad (4.70)$$

where W and ρ are, respectively, the weight adsorbed at pressure P and the liquid density of adsorbate, and c_1 a temperature-dependent constant. Thus, a plot of $\log W$ versus $[\log(P_0/P)]^2$ should yield a straight line with intercept $\log(V_m \rho)$, from which V_m is estimated. A modification of Eq. (4.70), suggested by Kaganer (1959),

is then used to estimate the surface of the micropores:

$$\log W = \log W_t - c_2 \left[\log \left(\frac{P_0}{P} \right) \right]^2, \quad (4.71)$$

where c_2 is another temperature-dependent constant. From the intercept of the straight line, resulting from a plot of $\log W$ versus $[\log(P_0/P)]^2$, W_t , the total weight adsorbed, is estimated. The total surface area is then computed using Eqs. (4.65) and (4.66). If the microporous adsorbent is very heterogeneous, then the exponent 2 in Eqs. (4.70) and (4.71) is replaced by a more general value m which may vary between 2 and 5.

Another widely-used method to obtain the pore size distribution of microporous materials is due to Horváth and Kawazoe (1983). They computed the potential energy profile for noble gas atoms adsorbed in a slit pore between graphitized carbon planes, separated by a distance ℓ . The adsorbed fluid was assumed to be a bulk fluid influenced by a mean potential characteristic of the adsorbent–adsorbate interactions. Horváth and Kawazoe (1983) developed the following correlation

$$\log \left(\frac{P}{P_0} \right) = \frac{N_a}{RT} \frac{c}{D_0^4(\ell - 2d_0)} \left[\frac{D_0^4}{3(\ell - d_0)^3} - \frac{D_0^{10}}{9(\ell - d_0)^9} - \frac{D_0^4}{3d_0^3} + \frac{D_0^{10}}{9d_0^9} \right]. \quad (4.72)$$

Here, $d_0 = (d_a + d_s)/2$, where d_a and d_s are, respectively, the diameters of the adsorbate and adsorbent, N_a is the number of adsorptive molecules per unit (in m^2) of adsorbent, C is a constant related to the number of atoms per unit area (in m^2) of adsorbent, the speed of light, the polarizabilities and magnetic susceptibilities of adsorbent and adsorbate, and $D_0 = (2/5)^{1/6} d_0$. Thus, corresponding to every relative pressure P/P_0 , there is a value of d_0 and, hence d_a . The effective pore diameter d_p is then given by $d_p = \ell - d_a$. The Horvath–Kawazoe relation was further refined by Saito and Foley (1995).

A third, and perhaps most accurate, method is the density-functional theory, the complete description of which is beyond the scope of this book (see Gelb *et al.*, 1999; Sahimi and Tsotsis, 2006). Here, we provide a brief description of the method. It should be clear to the reader that Eqs. (4.59) and (4.60) provide a means of computing the pore size distribution since they indicate that for every relative pressure P/P_0 , there is an effective pore radius r , a method that was first suggested by Barrett *et al.* (1951). A refined version of this method is based on using the adsorption isotherm since the primary adsorption isotherm does not depend on the connectivity of the pore space and, therefore, the isotherm can be thought of as the aggregate of the isotherms for the individual pores that make up the pore space. Thus, if r is the effective radius of a pore, one can write

$$N_a(P) = \int_{r_{\min}}^{r_{\max}} f(r) \rho(P, r) dr, \quad (4.73)$$

where $N_a(P)$ is the number of moles adsorbed at a pressure P , $r_{\min}$ and $r_{\max}$ are the effective radii of the smallest and largest pores present in the pore space, and $\rho(P, r)$

is the molar density of the adsorbed species at pressure P in a pore of radius r . $N_a(P)$ is directly measured, while one must use an equation of state to predict $\rho(P, r)$. Then, a numerical method is used to determine $f(r)$ from Eq. (4.73). This method was used successfully by Seaton *et al.* (1989), and later by Lastoskie *et al.* (1993). The density-functional theory is based on Eq. (4.73) in which a molecular model or atomistic simulation is utilized to estimate $\rho(P, r)$.

4.13.5

Pore Network Models

Computer simulation of sorption processes in porous media is similar to, but simpler than, mercury porosimetry described earlier in this chapter. As usual, we represent the pore space by a network of interconnected pores and throats. At the end of primary adsorption and at the maximum pressure, all pores of the network are filled by the condensate, for example, liquid nitrogen. This is, for example, the case for porous media that gives rise to hysteresis loops of types H1 and H2. As the pressure decreases, the condensate vaporizes from some of the pores adjacent to the external surface of the network. As described above, this is the linear and nearly horizontal part of the desorption curve shown in Figure 4.16. After the liquid in the pores at the surface has vaporized, some of the internal pores gain access to the vapor phase. Therefore, with decreasing pressure, the gas phase becomes sample-spanning.

It should, therefore, be clear that desorption is a percolation process. Let us simplify the problem for the moment and ignore the pore bodies. Then, the analog of p , the probability that a bond is open in percolation, is the fraction Y of the throats in which the adsorbate is below its condensation when the adsorbate would vaporize if all such throats had access to the vapor phase. Therefore, $(1 - Y)$ is the fraction of the throat that would contain liquid if all the throats had access to the vapor phase. The analog of the accessible fraction X^A is the fraction of the throats from which adsorbate has actually vaporized. That is, $(1 - X^A)$ is the fraction of the throats that contain the adsorbate in the liquid-like state.

Thus, a network model for simulating primary desorption (ignoring the pore bodies) is as follows. First, effective radii are selected from a pore size distribution and attributed to the throats. As the pressure is lowered, we examine the throats to see whether the adsorbate can vaporize in them based on the two criteria described above. The simulation continues until a sample-spanning cluster of the vapor-filled throats is formed. Typical results obtained with a simple-cubic network are shown in Figure 4.17, which have a striking similarity to those shown in Figure 4.16. The secondary desorption can also be simulated by a similar model, except that the simulation starts with a network in which a fraction Y_v of the throats are already occupied by the vapor phase, which have remained in the network at the end of secondary adsorption. Clearly, one may assume a pore size distribution with a few adjustable parameters and try to fit them by adjusting them such that the simulation results agree with the data. Various versions of this basic model have been used by several groups (Wall and Brown, 1981; Mason, 1982, 1983, 1988; Neimark,

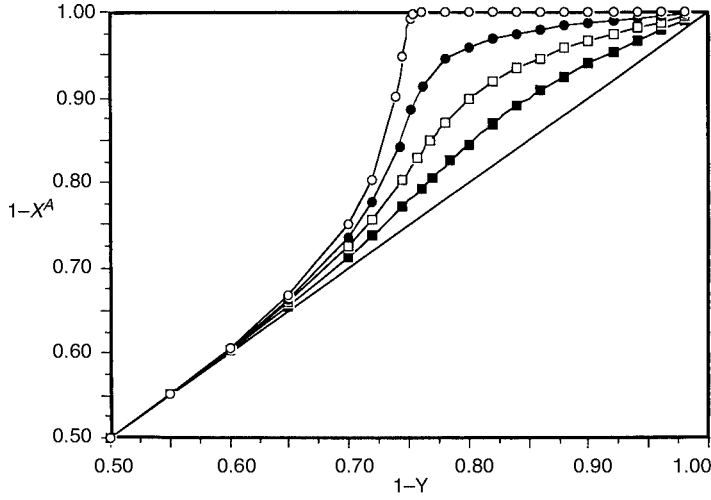


Figure 4.17 Sorption isotherms computed using the percolation-network model (after Liu *et al.*, 1993; courtesy of Professor N.A. Seaton).

1984b; Zhdanov *et al.*, 1987; Parlár and Yortsos, 1988, 1989; Ball and Evans, 1989; Mayagoitia *et al.*, 1989a; Zgrablich *et al.*, 1991; Liu *et al.*, 1993). A molecular pore network model that takes into account the interactions between the adsorbates and adsorbents was recently developed by Rajabbeigi *et al.* (2009a,b).

4.13.6

Percolation Models

Similar to mercury porosimetry, one can derive analytical formulae for the size distributions of the pores and throats occupied by the vapor or liquid phase during adsorption and desorption. Consider, for example, primary desorption during which a pore filled with liquid vaporizes if its radius $r > r_a$ is large enough, and if it is accessible to the vapor phase, where r_a is the radius that one computes based on the Kelvin equation or its modification, Eqs. (4.59) or (4.60), for a given P/P_0 . The fraction of the pores that are actually occupied by the vapor is given by

$$Y_i = X_i^A, \quad (4.74)$$

where X_i^A is the usual percolation accessibility function, and i denotes a site or a bond. The size distribution of the liquid-filled pores is simply given by

$$f_{Li}(r) = \begin{cases} \frac{f_i(r)}{(1 - Y_i)}, & r < r_a \\ f_i(r) \frac{(1 - \frac{Y_i}{P_i})}{(1 - Y_i)}, & r > r_a \end{cases} \quad (4.75)$$

which is similar to Eq. (4.56). Here, the quantity p_i ,

$$p_i = \int_{r_a}^{\infty} f_i(r) dr, \quad (4.76)$$

is simply the fraction of the pores or throats with $r > r_a$.

4.14

Pore Size Distribution from Small-Angle Scattering Data

As described earlier in this chapter, in order to use mercury porosimetry data or sorption isotherms to obtain the pore size distribution, one must have some information on the connectivity or the average coordination number $\langle Z \rangle$ of the pore space. In this section, we describe a method which is independent of $\langle Z \rangle$, and is based on small-angle scattering data, either small-angle X-ray scattering (SAXS), or small-angle neutron scattering (SANS). One measures the scattering intensity $I(q)$, where q is the magnitude of the scattering vector given by

$$q = 4\pi\lambda^{-1} \sin\left(\frac{\theta_s}{2}\right). \quad (4.77)$$

Here, λ is the wavelength of the radiation scattered by the sample through an angle θ_s . One then assumes a pore shape, for example, a sphere, a cylinder or a sheet-like structure with an effective size (for example, its radius) r and a number density n_p . Then, according to Vonk (1976), one has

$$I(q) = \bar{\rho}^2 \sum_{i=1}^n n_p V_p^2 |S_F(qr)|^2, \quad (4.78)$$

where V_p is the volume of a pore of effective radius r . Here, $\bar{\rho}$ is the difference in scattering amplitude densities of the solid matrix and the pore space, and $S_F(qr)$ is a *form factor* that depends on the pores' shape. For pores of any shape, one must have $S_F \sim 1$ as $q \rightarrow 0$, and $S_F \rightarrow 0$ for sufficiently large q . Thus, one measures $I(q)$, assumes a pore shape, fits the measurements to Eq. (4.78), and calculates n_p by a constrained least-squares fit.

Using this technique, and SAXS and SANS data, Hall *et al.* (1986) measured the pore size distributions of eight different rock samples, three of which were fractured, while two of them were sandstone. Figure 4.18 shows their results obtained with the SANS data and compared with those obtained with mercury porosimetry and sorption isotherms. The rock studied was a shale outcrop from Eastern Kentucky with porosity $\phi = 0.04$. In general, the pore size distributions obtained by the scattering methods tend to agree with those that secondary adsorption isotherms yield. Note that the sorption isotherms exhibit significant hysteresis, resulting in significantly different cumulative pore volumes, and that the results obtained with mercury porosimetry are in between the sorption results.

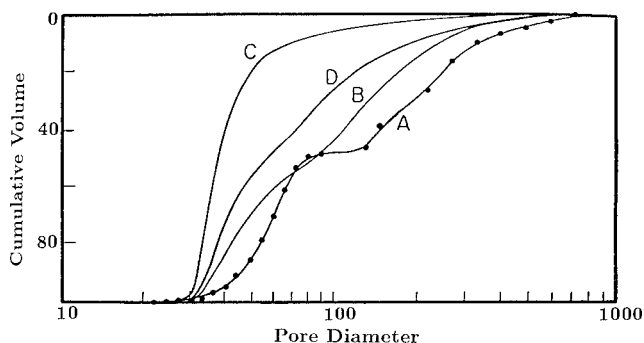


Figure 4.18 Comparison of cumulative pore size distribution of an oil shale obtained by the SANS (A) with those obtained by nitrogen adsorption (B), desorption (C), and mercury porosimetry (D) (Hall *et al.*, 1986).

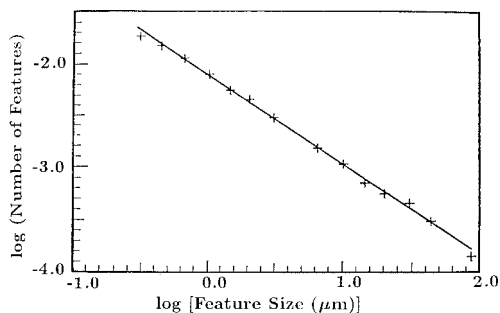


Figure 4.19 Fractal plot for Berea sandstone (after Krohn and Thompson, 1986).

Figure 4.19 also reveals a basic dilemma for anyone measuring the pore size distribution of a porous medium: Which method of measuring the pore size distribution should one use and when? How can one know *a priori* which method yields the most accurate results? Such questions, despite their significance, have not yet found definitive answers. While both mercury porosimetry and sorption methods suffer from the fact that *a priori* knowledge of the connectivity of the pore space and pore shapes is essential to their success, the scattering method also has its own shortcomings that (1) it contains the unknown shape factor S_F that depends on the pore shape, and (2) even if the pore shape is specified or known, the resulting pore size distribution may be sensitive to the shape. The conclusion is that all the methods of determining the pore size distribution that have been described so far have their own strengths and weaknesses.

4.15

Pore Size Distribution from Nuclear Magnetic Resonance

In a pioneering work, Brownstein and Tarr (1979) used the NMR method to study proton-spin relaxation in water in biological cells, and delineated the separate influ-

ence of diffusion and surface relaxivity (see below). The application of the NMR to determining the pore-size distribution of a porous medium seems to have been suggested first by Cohen and Mendelson (1982).

In the NMR method, the porous medium is first saturated with a suitable fluid such as water. Then, proton nuclei are aligned in a certain direction by a strong magnetic field. An appropriate pulse is then applied and the magnetization relaxation is measured as a function of the time. Magnetization relaxation is caused by the interaction of the pores' surface with the fluid near the surface as well as with that in the bulk. Therefore, the relaxation rate provides direct information about the surface-to-volume ratio and, hence, an effective pore size. If the porous medium is characterized by a pore size distribution, and if there are regions of the pore space that are separated by more than one diffusion length (by which the molecules move in the pore space), such regions can be distinguished in the relaxation data. If the pore space of the medium is too complex, the NMR relaxation may not be able to reveal all of its complexities. Moreover, if the signal-to-noise ratio is finite, extracting a pore size distribution may be too difficult or be subject to large errors. Despite such difficulties, the NMR relaxation has been used for probing the pore space of various types of porous media and obtaining their pore size distributions. Let us now describe how the NMR data are analyzed for determining the pore size distribution by following Cohen and Mendelson (1982) and Schmidt *et al.* (1986).

One assumes that each pore contains two types of fluid. One is a layer of thickness l_a adsorbed on the pore surface with relaxation time t_a , while the other is the fluid in the bulk away from the surface with relaxation time t_b . In the presence of a field applied from the external surface, $t_a < t_b$ because the applied field hinders diffusion of the fluid. The ratio t_a/t_b depends on the nature of the adsorbent and the surface geometry. The NMR relaxation, together with diffusion, act to smooth out any spatial gradient in the magnetization that exists between the adsorbed and bulk fluids as well as between fluids in adjacent pores. The governing equation for the magnetization M_z is given by

$$\frac{\partial M_z}{\partial t} = \sigma_p (\mathbf{M} \times \mathbf{H})_z - \frac{M_z - M_\infty}{t_r} + D \frac{\partial^2 M_z}{\partial z^2}, \quad (4.79)$$

where $\mathbf{H}$ is the magnetic field, σ_p is the proton gyrometric ratio, D is the diffusivity, t_r is a relaxation time, and M_∞ is the equilibrium magnetization.

In a pore of effective radius r and length l , the magnetic field gradients between the surface and the bulk are smoothed by diffusion in a time

$$t_d = \left(\frac{r^2}{6D} \right) \left(\frac{S_p l}{V_p} \right), \quad (4.80)$$

where S_p and V_p are the surface and pore volume, respectively. Each pore is characterized by a relaxation time t_p . If $t_d < t_p$, then there will be an averaged signal for that pore. If, however, $t_d > t_p$, then there will be a complex signal caused by the spatial inhomogeneities. Thus, there is a critical pore radius r_c such that, if $r < r_c$, one will observe an averaged signal. However, for $r > r_c$, there will be a complex

or multicomponent signal. For pores with $r < r_c$, the average relaxation time $\langle t \rangle$ is given by

$$\frac{1}{\langle t \rangle} = \frac{1 - \frac{S_p l}{V_p}}{t_b} + \frac{\frac{S_p l}{V_p}}{t_a}, \quad (4.81)$$

and, therefore, by measuring $\langle t \rangle$ for a given pore, one obtains an estimate of S_p/V_p .

So far, we have considered diffusion only within a pore. One should also consider diffusion *between* the pores that depends on the distance L_p between them. For a porous medium of spherical pores of radius r , one has $L_p \sim r\phi^{-1/3}$, where ϕ is the porosity. If diffusion between the pores totally dominates the porous medium's response, only one relaxation time is observed for the entire medium. Normally, however, diffusion between pores is not significant, and the porous medium behaves as a collection of isolated pores. In this situation, each pore has its own relaxation time that depends on its surface-to-volume ratio. Thus, if one groups pores of the same effective radius together, one can write

$$M_z(t) = M_\infty + (M_0 - M_\infty) \int_{\omega_{\min}}^{\omega_{\max}} P(\omega) \exp(-\omega t) d\omega, \quad (4.82)$$

where $\omega = t_\tau^{-1}$ is the frequency of the relaxation, and $P(\omega)$ is the fraction of the fluid that resides in pores with relaxation frequency ω . Equation (4.82) is rewritten as

$$M_z(t) = M_\infty - \int_{\omega_{\min}}^{\omega_{\max}} P_1(\omega) \exp(-\omega t) d\omega, \quad (4.83)$$

where $P_1(\omega) = (M_\infty - M_0)P(\omega)$. Because one measures $M_z(t)$ at various discrete times, τ_j ($j = 1, 2, \dots, N$), Eq. (4.83) is written in a discretized form

$$M_z(\tau_j) = M_\infty - \sum_{\omega_i = \omega_{\min}}^{\omega_{\max}} \exp(-\omega_i \tau_j) P_1(\omega_i). \quad (4.84)$$

Note that $P(\omega)$ is normalized and, therefore, $\sum_{\omega_i = \omega_{\min}}^{\omega_{\max}} P(\omega_i) = 1$. Equation (4.84) is then solved for $m+2$ unknowns and N data points, with the interval $(\omega_{\min}, \omega_{\max})$ divided into m subintervals of length $\Delta\omega = (\omega_{\max} - \omega_{\min})/m$. If $N \geq m+2$, then Eq. (4.84) is used to estimate S_p/V_p for pores with frequency ω_i . Assuming a pore shape, its effective size is estimated.

The NMR method is based on the assumption that diffusion between pores is not important and, hence, the pores can be treated independently. Cohen and Mendelson (1982) and Mendelson (1982) discussed the conditions under which the assumption of independence of the pores is valid. One geometrical requirement for the validity of this assumption is that the throats must be relatively narrow, because then diffusion between pores will be severely restricted, which is certainly valid for some, but not all, porous media. In the latter case, one can still obtain a

pore size distribution, but the effective sizes that are obtained are only rough estimates of the true values. Since a pore shape must be assumed anyway, which is an approximation by itself, the calculated pore size distribution will be based on two approximations. Latour *et al.* (1992) presented some data on the temperature-dependence of decay of the spectra as evidence for the assumption of isolated or uncoupled pores. McCall *et al.* (1991) utilized the method of Cohen and Mendelson (1982), and demonstrated how the spectrum of decay narrows as the diffusivity increases. Mendelson (1986) extended the above analysis to porous media with fractal properties (see below).

Schmidt *et al.* (1986), Lipsicas *et al.* (1986), and Billardo *et al.* (1991) used the NMR technique to measure the pore size distributions of various sandstones. Schmidt *et al.* (1986) compared their results with those obtained by mercury porosimetry and showed that the NMR technique is more sensitive to the details of the pore structure, and can also reveal a bimodal pore size distribution if there is one. Strange and Webber (1997) used the NMR to determine the median pore size and pore size distribution of a variety of porous media. Aksnes *et al.* (2001) used the method to measure the pore size distributions of a series of mesoporous silica materials. Minagawa *et al.* (2007) used the NRM method for characterizing sand sediments by their pore-size distribution and effective permeability. A good review of the applications of the NRM method to characterization of carbonate rocks is given by Westphal *et al.* (2005).

4.16

Determination of the Connectivity of Porous Media

One of the simplest concepts for characterizing the topology of a porous medium is the coordination number Z which is loosely defined as the number of throats that meet at a pore of the medium. For an irregular pore space, one must define an average coordination number $\langle Z \rangle$, and the average must be taken over a large enough sample. For microscopically-disordered, macroscopically-homogeneous media, $\langle Z \rangle$ is independent of sample size. Moreover, topological properties of porous media are invariant under any deformation of the pore space and solid matrix.

How can one estimate $\langle Z \rangle$ and other topological properties of a porous medium? Stereology (Underwood, 1970) and serial sectioning (Pathak *et al.*, 1982; Lin and Hamasaki, 1983; Koplik *et al.*, 1984; Yanuka *et al.*, 1984; Lin *et al.*, 1986; Kwiecien *et al.*, 1990; Tsakiroglou and Payatakes, 2000) were used in the past to deduce the 3D structure of porous media. In particular, Kwiecien *et al.* (1990) developed computer programs that take data, analyze them, and generate the computer image of a porous medium and its various properties, for example, the pore and throat size distributions and the average coordination number. However, neither of the two methods is used routinely at present. More popular are indirect methods by which only *statistical* information about the structure of the system is obtained. Some of the indirect methods are the NMR, porosimetry, and sorption experiments, which may yield parts or all of the pore size distribution, and if $\langle Z \rangle$ is treated as an adjustable parameter, it can also simultaneously be estimated with the pore size

distribution. Mason (1988) and Seaton (1991) (see also Liu *et al.*, 1992 for further elaboration) developed a direct method for estimating $\langle Z \rangle$ which is based on a percolation model of sorption phenomena. In what follows, we describe Seaton's method.

Seaton's method is based on finite-size scaling analysis described in Chapter 3, according to which

$$X^A(p) = L^{-\frac{\beta}{\nu}} f \left[(p - p_c) L^{\frac{1}{\nu}} \right], \quad (4.85)$$

which is rewritten as

$$\langle Z \rangle X^A(p) = L^{-\frac{\beta}{\nu}} f \left[(\langle Z \rangle p - B_c) L^{\frac{1}{\nu}} \right], \quad (4.86)$$

using $B_c = \langle Z \rangle p_{cb}$ (see Chapter 3). Accurate estimates of $X^A(p)$ were obtained by Kirkpatrick (1979) for a simple-cubic network for various sizes L , and can be shown to follow Eq. (4.85).

Consider, as an example, the H2 loop in Figure 4.15. The desorption curve has three segments indicated by 1, 2 and 3 in the figure. In the 1–2 interval, the isotherm is almost linear, and occurs due to the decompression of the liquid adsorbate (for example, nitrogen) in the pores. In the corresponding percolation network, Y , the fraction of open pores (those in which the pressure is below the condensation pressure) increases, but $X^A(Y)$, their accessible fraction, is still zero because a sample-spanning cluster of open pores has not been formed yet. At point 2, the network reaches its percolation threshold, a sample-spanning cluster of open pores is formed, and the metastable liquid in the pores of the cluster vaporizes. If one decreases the pressure further, the number of pores containing metastable adsorbate, and the number of pores whose liquid content has vaporized, both increase. At point 3, almost all the pores in which the pressure is below the condensation pressure can lose the adsorbate (nitrogen) by vaporizing it and, therefore, $X^A(Y) \simeq Y$. Note that in a *finite* percolation network, one has a *smear out* transition in which the discontinuity in the desorption isotherm causes a rapid increase in the slope. A similar analysis may be used for interpreting the H1 loop. Thus, Seaton's method consists of two steps:

1. $X^A(Y)$ is determined from the sorption data, and
2. $\langle Z \rangle$ and L are determined by fitting Eq. (4.86) to the $X^A(Y)$ data.

Similar to most of the methods of determining the pore size distribution described above, it is necessary to assume a relation between the pore radius and length. For example, one may assume that the length and the radius of a pore are uncorrelated. Note that $X^A(Y)/Y$, which is the ratio of the number of pores in the percolation cluster and the number of pores below their condensation pressures, can also be written as N_p/N_b , where N_b is the number of moles of adsorbate that would desorb if all the pores containing it below its condensation pressure had access to the vapor phase, and N_p is the number of moles of adsorbate that actually have desorbed at

that pressure. If N_A is the number of moles of adsorbate that are present in the pores at a given pressure during the adsorption experiment; N_D is the number of moles of adsorbate that are present in the pores at that pressure during the desorption experiment, and N_F is the number of moles of adsorbate that would have been present in the pores at that pressure during the desorption experiment, had no adsorbate vaporized from the pores that contain the adsorbate below its condensation pressure. Then, $N_p = N_F - N_D$, and $N_b = N_F - N_A$ and, therefore,

$$\frac{X^A(Y)}{Y} = \frac{N_F - N_D}{N_F - N_A}, \quad (4.87)$$

so that $X^A(Y)/Y$ is expressed in terms of measurable quantities. The final step is to determine Y , so that $X^A(Y)$ can be estimated from Eq. (4.87). If $f(r)$ is the normalized distribution of numbers of throat radius r , then, for a given pressure, one has

$$Y = \int_r^\infty f(x) dx, \quad (4.88)$$

where r is the throat radius in which the adsorbate condenses at the specified pressure. Therefore, given the distribution $f(r)$ determined from mercury porosimetry, sorption isotherms, or any other method, Y and, hence, $X^A(Y)$ are estimated.

Seaton's method, while very useful, is not free of problems. Rigby (2000) applied Seaton's method to the nitrogen sorption data for both whole (particle size of about 3 mm) and fragmented (particle size of about 30 μm) samples of unimodal, mesoporous, alumina and silica catalyst support pellets. His analysis indicated that the size L of the equivalent networks for both cases was about the same, even though the actual particle sizes differed by about two orders of magnitude. This seemingly contradictory result was explained by observing that the pore structure in the materials was not random, but contained significant correlations. Therefore, regions with pores of more or less similar sizes could behave effectively as a single pore. If the extent of such regions is large, then Eqs. (4.86) and (4.87) will underestimate the true length L of the equivalent pore network of a porous sample. For such cases, a hierarchical network model, described in Section 4.12.6, may be more appropriate than a simple random pore network model. In addition, Meyers *et al.* (2001) attempted to match nitrogen desorption isotherms for silica particles with simulated isotherms computed with the network models with *variable* network size, connectivity, and spatial distribution of pore sizes with reasonable success. Murray *et al.* (1999), on the other hand, combined mercury porosimetry and nitrogen adsorption measurements in order to probe the connectivity of a porous sample. Mason (1988)'s method has many similarities with Seaton's, except that he adopted the Bethe lattice as the network model of the pore space.

A more precise method of characterizing the connectivity of a pore space relies on the *Betti numbers*, quantities that were described by Barrett and Yust (1970) for metallurgical systems, and by Lin and Cohen (1982) and Pathak *et al.* (1982) for porous rock (see also Tsakiroglou and Payatakes, 2000). A fundamental theorem of

topology (see, for example, Alexandroff, 1961) states that *two structures are topologically equivalent if and only if their Betti numbers are all equal*. For a given structure, one may define many Betti numbers, and their precise definition requires considerable knowledge of topology. For our present purpose though, we only need the first three Betti numbers.

The zeroth Betti number β_0 is the number of isolated clusters in a structure. In other words, β_0 is the number of *separate* components that make a structure. For example, the grain space of a single, finite sandstone has $\beta_0 = 1$. Thus, $\beta_0 > 1$ may indicate that the structure contains isolated porosity.

The first Betti number β_1 is the number of holes through a structure, or the maximum number of non-intersecting closed curves that can be drawn on the surface of the structure *without separating it*. It is given by $\beta_1 = N_e - N_s + 1$, where N_e is the number of edges, and N_s is the number of sites (vertices) of the network equivalent of the pore space. For example, if a torus is cut along a closed curve, the resulting solid can be deformed into a cylinder, whereas if the cylinder is cut along a closed curve, it separates into two disconnected clusters. Thus, the first Betti number of a torus is one, whereas that of a cylinder is zero.

The notion of the *genus* of a surface is also used for characterizing the topology of a complex system. Also called *holeyness*, the genus G and the first Betti number are equal for certain graphs lying on surfaces. One can use a genus per unit volume G_V by normalizing it over the volume in which it is measured. For large systems, $\beta_1 \simeq E - N_V$ and, therefore, $G_V = \beta_1/N_V = (E/N_V) - 1$. Note that for graphs or a network equivalent of a porous medium, G_V is half of the coordination number, but the notions of genus and genus per unit volume are more general than the coordination number. It is clear that the first Betti number, or genus, is also a measure of multiplicity of independent paths in a structure.

The second Betti number β_2 is a measure of the sidedness of a structure. For example, a solid structure containing n isolated pores is n -sided. Note that the Betti numbers may also be defined for *both* the solid matrix, β_0^s, β_1^s , and β_2^s , and for the pore space, β_0^p, β_1^p , and β_2^p , but they are related through the following relationships

$$\begin{aligned}\beta_0^p &= 1 + \beta_2^s, \\ \beta_1^p &= \beta_1^s = G, \\ \beta_0^s &= 1 + \beta_2^p,\end{aligned}\tag{4.89}$$

and, therefore

$$\beta_0^p + \beta_2^p = \beta_0^s + \beta_2^s,\tag{4.90}$$

implying that the topologies of pore space and solid matrix are *conjugate*, and one needs to only measure one of them. For microscopically-disordered porous media, the Betti numbers must be averaged over a large enough sample. Although, as mentioned above, one may also use topological measures per unit volume, such measurements suffer from the disadvantage that they depend on the unit chosen for the volume. For example, a heavily-consolidated rock with many large, irregular grains that have many contacts with one another may have the same genus per unit

volume as a lightly-consolidated rock that consists of small, well-rounded grains with few grain-to-grain contacts.

Topology and geometric shapes are related through the Gauss–Bonnett theorem (see, for example, Kreyszig, 1959). The local Gaussian curvature of a surface, C_G , is given by $C_G = C_1 C_2$, where C_1 and C_2 are the local principal curvatures of the surface. C_G is negative if the surface is saddle-shaped and positive if it is convex or concave. One defines the integral Gaussian curvature $\langle C_G \rangle$ by

$$\langle C_G \rangle = \int_s C_G ds. \quad (4.91)$$

According to the Gauss–Bonnett theorem, one has

$$\langle C_G \rangle = 4\pi(1 - G_V). \quad (4.92)$$

Natural rock is highly porous and has large genus. It also has large negative $\langle C_G \rangle$. Therefore, it must be riddled with pore wall areas that are saddle-shaped.

Such topological properties were measured by Pathak *et al.* (1982) for artificial porous media. They sintered three different copper powders: (1) spherically shaped grains in the range 30–90 μm ; (2) electrolytically prepared grains of less regular shape in the range 30–90 μm , and (3) electrolytically prepared grains in the size range 250–300 μm . The sintering process parallels, in many important aspects, the diagenesis of sedimentary rock. With increasing sintering, the initial rough surface and edges of the original powders are smoothed out; the surface areas per unit volume no longer depend on the original shape and exhibit a universal dependence on ϕ . By using cold compression of spherical grains, Pathak *et al.* also prepared polyhedral-shaped particles. Using serial sectioning, they measured the genus per

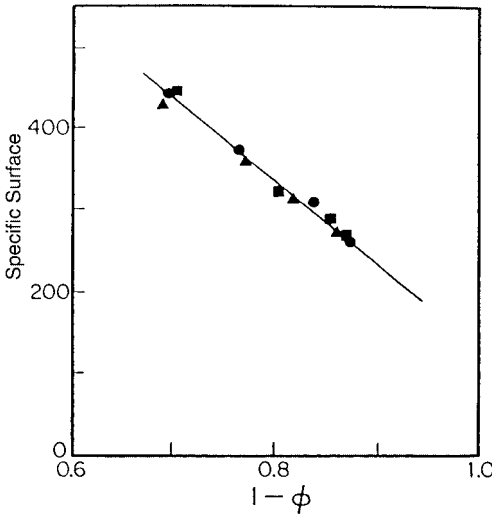


Figure 4.20 Dependence of the specific surface on the porosity ϕ of a sintered copper powder. The data are for spherical (triangles), polyhedra (squares), and irregular (circles) particles (after Pathak *et al.*, 1982).

unit volume G_V and surface area per unit volume Ξ of their porous media. Figure 4.20 presents the variations of Ξ with ϕ .

Lin and Cohen (1982) studied six different Berea sandstone samples and measured several of their topological properties by serial sectioning and image analysis. β_1 was measured only for the main pore subsystem, with its minimum and maximum values being 91 and 280, while β_0 was found to be about 23, indicating a large amount of isolated porosity. The number of contacts per pore section which had a broad distribution were also measured. The connectivity of the pore or grain system of the Berea sandstone was found to be lower than those of regular monosized sphere packs with similar porosities and the same mean grain diameter. To summarize, most studies indicate that for sandstones, an average coordination number between four to eight is a reasonable estimate.

4.17

Fractal Properties of Porous Media

Thus far, we have described the pore size distribution and connectivity of various types of porous media as well as their measurement and modeling. The average coordination number of sedimentary rock varies anywhere from four or five to 15. Many other types of porous media, for example, catalyst particles, coals, and membranes, may also have an average coordination number in the same range. Therefore, what distinguishes natural porous media from other types of porous materials is their geometry, that is, the shapes and sizes of their pores. Another factor that helps distinguishing various classes of porous media is the *fractal* properties of the pore space. In Section 3.8, we described the self-similar and fractal properties of percolation networks. It has been shown by several research groups over the last three decades that many types of porous media and materials exhibit fractal and self-similar properties, and characterization of such properties attracted considerable attention. Many theoretical, computer simulation, and experimental studies were undertaken in an attempt to understand such properties of porous media. In this section, we describe and review fractal properties of porous media, while those of fractures and fractured porous media will be described in Chapter 6. There are many methods of measuring the fractal properties, and what follows is a description of each.

4.17.1

Adsorption Methods

A powerful technique for measuring fractal properties of porous materials is based on adsorption. A comprehensive review of the subject was given by Pfeifer and Liu (1996), whom we follow in this section. As discussed in Section 3.8, the fractal dimensions measure the space-filling ability of a system. Since adsorption takes place on a surface, one must find a way of characterizing the space-filling ability of adsorptive surfaces. Clearly, a smooth surface provides smaller area for adsorption

than a rough one with many “hills” and “valleys.” The most precise function for characterizing the space-filling ability of a surface is $V(z)$, the volume of all points outside the solid surface at a distance less than or equal to z . Then, $I_z = dV/dz$ is the area of the interface (film), and we refer to $f(S, I_z)$ as film of thickness z , where S represents the solid surface. A self-similar surface with *surface fractal dimension* D_s is one for which

$$\frac{dV}{dz} \propto z^{2-D_s}, \quad l_{\min} \leq z \leq l_{\max}, \quad (4.93)$$

where $l_{\min}$ and $l_{\max}$ are the lower and upper cutoff scales for fractality of the surface. Equation (4.93) indicates that the film area on a fractal surface decreases with increasing film thickness (by filling the “vallies”). If the fractal regime starts at the smallest scale, namely, at the monolayer scale where the diameter of an adsorbed molecule is a_0 , then one may integrate Eq. (4.93). With N_t being the total number of molecules in a monolayer, one obtains

$$V(z) = \begin{cases} N_t a_0^3 \left(\frac{z}{a_0}\right)^{3-D_s}, & 2 \leq D_s \leq 3, \\ N_t a_0^3 \left[1 + \ln\left(\frac{z}{a_0}\right)\right], & D_s = 3, \text{ non-uniform space filling}, \\ N_t a_0^3, & D_s = 3, \text{ uniform space filling}, \end{cases} \quad (4.94)$$

where $a_0 \leq z \leq l_{\max}$. Equation (4.94) compares surfaces with variable D_s and fixed number of adsorption site, N_t . In the surface-area consideration, one compares surfaces with variable D_s and fixed diameter L (the largest distance between two points on the surface). To switch from one to the other, one must use the relation

$$N_t = \left(\frac{L}{l_{\max}}\right)^3 \left(\frac{l_{\max}}{a_0}\right)^{D_s}. \quad (4.95)$$

The first factor in Eq. (4.95) counts how many fractal, identical “pieces” can cover the surface, while the second factor accounts for the number of surface sites on each piece. If the surface is fractal over the entire range of length scales in (a_0, L) ($l_{\max} = L$), then,

$$N_t = \left(\frac{L}{a_0}\right)^{D_s}. \quad (4.96)$$

When the chemical potential difference $\Delta\mu$, defined by Eq. (4.57), is very low, one has van der Waals wetting that is independent of the surface tension, and depends only on the surface potential (the surface chemical composition and structure). If, on the other hand, $\Delta\mu$ exceeds a critical value $\Delta\mu_c$, one obtains capillary wetting that does depend on the surface tension and was described earlier in this chapter. Therefore, one must also differentiate between adsorption on surfaces that are wetted by van der Waals wetting from those due to capillary wetting. An example of van der Waals wetting is the Frenkel–Halsey–Hill (FHH) isotherm on a flat surface

Table 4.2 Multilayer adsorption isotherms on fractal surfaces with surface fractal dimension D_s . c_1 and c_2 are constant (adopted from Pfeifer and Liu, 1996).

Range of $\Delta\mu$	van der Waals wetting $\Delta\mu \leq \Delta\mu_c$	Capillary wetting $\Delta\mu \geq \Delta\mu_c$
$D_s \rightarrow 2$	Range increases	Range vanishes
$D_s \rightarrow 3$	Range decreases	Range increases
Isotherms		
$2 \leq D_s \leq 3$	$N \propto [-\ln(P/P_0)]^{-(3-D_s)/3}$	$N \propto [-\ln(P/P_0)]^{-(3-D_s)}$
$D_s = 3$	$N \propto c_1 - \ln[-\ln(P/P_0)]$	$N \propto c_2 - \ln[-\ln(P/P_0)]$
Interactions	Substrate potential pulls liquid–gas interface to the surface; film grows slowly	Surface tension pushes liquid–gas interface from the surface; film grows rapidly

which was already described earlier. The critical chemical potential difference $\Delta\mu_c$ is the maximum of $\Delta\mu$ when it is plotted versus the distance z .

Using Eqs. (4.94) and (4.95), one can derive (Pfeifer and Liu, 1996) the adsorption isotherms on fractal surfaces for both the van der Waals and capillary wettings. The results are listed in Table 4.2. Note that the FHH isotherm, from Eq. (4.69), is given by

$$N \propto \left[-\ln \left(\frac{P}{P_0} \right) \right]^{-\frac{1}{\zeta}}, \quad (4.97)$$

where ζ is a constant. Therefore, the isotherms listed in Table 4.2 for $2 \leq D_s \leq 3$ represent generalization of the FHH isotherm to non-smooth and fractal surfaces.

Using gas adsorption and Eq. (4.96), Avnir *et al.* (1983) measured pore surface properties at the nanometer scale. The implicit assumption is that surface coverage is *uniquely* determined by the adsorbed gas species. Equation (4.96) is usually referred to as the *box-counting method* of determining the fractal dimension. Avnir *et al.* (1983, 1985) extended the range well beyond molecular sizes by studying sorption properties of fractal surfaces in larger particles, and by considering their scaling with the particles' Euclidean size R using Eq. (4.96) and a single species. The following equation holds

$$N \sim R^{D_s-3}, \quad (4.98)$$

if we assume that the surface area is proportional to R^{D_s} , and that the particle weight varies with the volume as R^3 . To measure D_s , the system under study is sieved into several fractions. For each fraction, the apparent monolayer value of N is determined by any convenient method, for example, adsorption from solution. If D_s is very close to three, which is indicative of very wiggly porous material, then N becomes independent of R . One may also express Eq. (4.98) in terms of an *apparent* or effective surface area that is simply proportional to N , as given by Eq. (4.98).

Adsorption can also determine the range of self-similar and fractal behavior. If a surface fractal dimension D_s is estimated from the measurements of monolayer values of sieved fractions of particle diameters from $R_{\min}$ to $R_{\max}$, with a probe molecule of cross-sectional area A_c , and if, $A_{\max} = A_c(R_{\max}/R_{\min})^2$, then the range of self-similarity of the surface is between A_c and $A_{\max}$. It should be clear that in order to obtain the maximum amount of information about the geometry of a surface at the molecular scale, one should use molecular probes with surface coverages (per site) that are as small as possible. In practice, this is the case since nitrogen or argon is usually used.

Measurements of Avnir *et al.* (1983, 1985) revealed interesting results. Six carbonate rock samples were found to have a fractal pore surface with $2.16 \leq D_s \leq 2.97$, seven types of soils with $2.19 \leq D_s \leq 2.99$, and a number of crushed rock samples from nuclear test sites with fractal dimensions in the range, $2.7 \leq D_s \leq 3$. These estimates will be compared with those obtained by other methods described below. Estimates for D_s for many other types of surfaces and porous materials are listed by Pfeifer and Liu (1996).

Adsorption methods are *not* free of limitations or potential problems. If, as discussed by de Gennes (1985), chemical disorder on the pore surface is important, or if molecular conformation and orientation are functions of the structure of the pore surface, then adsorption yields *biased* estimates of D_s . Moreover, if D_s is close to three, which is indicative of a highly rough surface, some parts of the surface *shadows* the neighboring surfaces leading to incomplete adsorption and a lower bound to D_s , rather than its true value, but for various types of porous media adsorption methods yield estimates of D_s that are in general agreement with those obtained by other methods. One other shortcoming of the adsorption methods is that the range of the adsorbates' size is very narrow, usually from 0.2 to 1 nm.

4.17.2

Chord-Length Measurements

There are two basic methods of measuring chord lengths, namely, on fracture surfaces and on thin sections. What follows is a description of each method.

4.17.2.1 Chord-Length Measurements on Fracture Surfaces

A detailed description of chord-length measurements on fracture surfaces is given by Krohn and Thompson (1986) and Krohn (1988a), which we summarize here. At the outset, however, we should mention that they did not distinguish between a fractal pore surface with fractal dimensionality D_s and a fractal pore space with fractal dimensionality D_f . Katz and Thompson (1985) argued that for sandstones, $D_s = D_f$. This issue will be discussed shortly.

With this method, one counts features in a large number (1000 or more) of horizontal lines across a digitized image of a fracture surface (see Figure 4.21). The counting is then repeated for a number of magnifications and locations. One begins by selecting a highly structured location on the surface and digitizing the images at several different magnifications. A constant resolution for feature detection

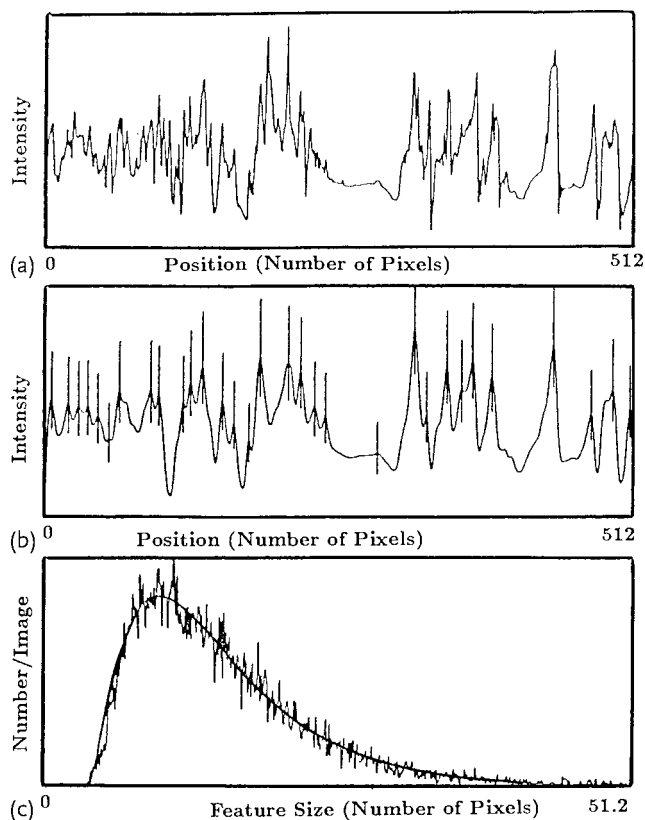


Figure 4.21 The size distribution of surface features of a sandstone; (a) and (b) show two different samples, while (c) presents the number distribution of the features versus their size (after Krohn and Thompson, 1986).

is then set using a digital low-pass filter. Feature sizes, defined as the distance between local maxima, are then measured, based on which a histogram is generated which is then linearized and placed on a log-log plot. That this procedure can be carried out is due to the probability of detecting a feature at each magnification being known. The effect of various factors on the construction of the histograms and the resulting plots were thoroughly investigated by Krohn and Thompson (1986) and Krohn (1988a).

This technique does not depend on the delineation of the pore or grain space. It is an automatic method that statistically measures structural features using scanning electron microscopy (SEM) images of the surface. A change in contrast in the secondary electron intensity of the SEM, which results in a local maximum in intensity, is defined as the edge of a feature. The technique makes it possible to decide whether features of a given size dominate the geometry of the pore space. Ehrlich *et al.* (1980) and Orford and Whalley (1983) also used the SEM measurements of grain roughness to analyze the results in terms of fractal concepts. However, they

measured the roughness of individual grains by analyzing the outline of the grains in a grain mount, whereas the fracture surface technique measures the pore-grain interface without isolating individual grains. As a result, while the fracture surface technique yields a single fractal dimension for *all* of the lengths, the fractal analysis of Orford and Whalley (1983) does not.

The next step is analyzing the feature distribution. For fractal behavior, the number of features $N_{\text{cm}}(l)$, counted per centimeter, for features of size l is expressed as

$$N_{\text{cm}}(l) \sim l^{2-D_f}, \quad (4.99)$$

where $l_{\min} \leq l \leq l_{\max}$ with $l_{\min}$ and $l_{\max}$ being the limits of fractal behavior. For $l > l_{\max}$, the samples are homogeneous and $D_f = 3$, which is the case if the geometrical features appear only as statistically random noise. Because all the measurements are made from images, one expresses the feature sizes in terms of pixels, where a pixel is 1/512 of the image. One obtains a sequence of intensities $I(J)$ for representing the digitized data, where $1 \leq J \leq 512$ is a pixel. If one edge of a feature is at J_1 and the other at J_2 , then the feature size l is $l = J_2 - J_1$. For each image, the width (in centimeters) of the field of view is $12/M$, where M is the magnification. Therefore,

$$N_{\text{cm}}(l) = a \left(\frac{12l}{512M} \right)^{2-D_f}. \quad (4.100)$$

However, the true number of features counted, $N(l)$ is given by

$$N(l) = N_{\text{cm}}(l) P_f(l) L(l), \quad (4.101)$$

where P_f is the probability of finding a feature, and $L(l)$ is the distance (in centimeters) over which the features are counted. The digital filter sets $P_f(l)$ that is determined by performing the Fourier transform of the impulse response and expressing the amplitude as a function of l . The probability of resolving a feature is directly dependent upon the filter's amplitude, and equals one at the largest feature sizes. P_f is set to zero for $l < l_0$, where the amplitude of the filter becomes less than the signal-to-noise threshold in order to simulate the amplitude threshold for the removal of the noise. The final expression for $N(l)$ is

$$N(l) = a P_f(l) \left(\frac{12l}{512M} \right)^{2-D_f} \left(\frac{12}{M} \right) [1 - F(l-1)], \quad (4.102)$$

where $F(l-1)$ is the fraction of the field of view occupied by features of size less than l ,

$$F(l-1) = \frac{1}{512} \sum_{i=1}^{l-1} i N(i), \quad (4.103)$$

and $N(i)$ is the number of features of size i . Thus, the model contains two adjustable parameters, namely, the prefactor a and the fractal dimension D_f .

The chord lengths that are measured by this technique can represent either pore-surface structure or fracture-surface structure. The method does have the drawback that the fracturing process may introduce unwanted structures. Thus, one must make sure that a section of the surface is measured, not a projection. Figure 4.21 shows typical results for Berea sandstone with a porosity of about 0.2 for which $D_f \simeq 2.85$, and $l_{\max} > 32 \mu\text{m}$. In general, after examining a dozen sandstone samples, it was found that $2.55 < D_f < 2.85$.

4.17.2.2 Chord-Length Measurements on Thin Sections

This method is not as accurate as the fracture-surface technique. It was essentially developed to provide data that are complementary to those obtained with the fracture-surface method. The method does, however, have its own advantages. For example, it may be used for measuring the porosity and its spatial distribution. Let us describe the method by following Krohn (1988a).

In this method, one digitizes SEM images and delineates the pore space whenever the intensity is less than a set gray level. Usually, the edge of a feature appears bright on the SEM images. If one examines the gray level histograms of the images, one finds that the distribution of grains always appears to be brighter than the pore distribution. The gray level for pore fill is between those of grains and pores and, therefore, it is important to measure the pores within the pore fill. Once the SEM images are digitized, chord lengths are measured from the interception of horizontal lines with the surface of pores. Using a logarithmic bin size, one constructs a histogram of the number of chords with lengths that are in a given range. The results are not dependent on the specific choice of the gray level as long as the method is consistent from magnification to magnification.

Typical results are shown in Figure 4.22 for Coconino sandstone with porosity of about 0.1. The estimated fractal dimension is about 2.75, close to that of Berea sandstone. In general, the estimates of D_f obtained with thin sections agree with

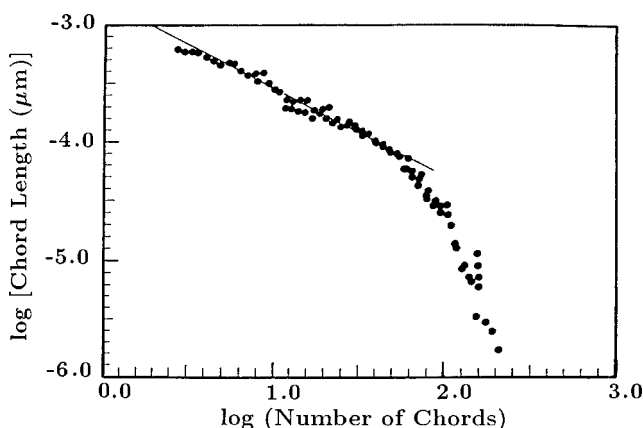


Figure 4.22 Fractal plot for Coconino sandstone, indicating the lower and upper limits of fractality (after Krohn, 1988a).

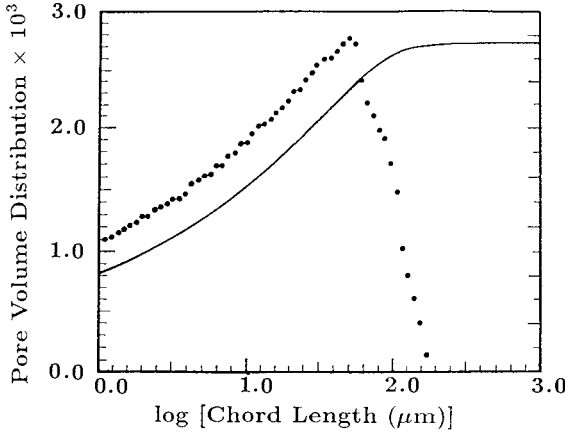


Figure 4.23 Pore volume distribution for Coconino sandstone, indicating the upper limit of fractal behavior (after Krohn, 1988a).

those obtained with the fracture surface technique. Note that both methods yield estimates for $l_{\max}$, the upper limit of fractal behavior. The chord-length methods do not contain any information on the correlation functions and, therefore, estimates of D_f obtained with the two methods are *not* unambiguous evidence for fractal behavior (see below).

The linear intersection of the pore space that one uses in chord-length measurements on thin sections is utilized for measuring a pore volume distribution defined as the porosity associated with each chord-length by

$$\phi(L) = N_C(l)l(\Delta l)^2, \quad (4.104)$$

where $N_C(l)$ is the number of chords per unit volume of length l , and $(\Delta l)^2$ is the cross-sectional area associated with each chord that is equal to one pixel. To estimate $N_C(l)$, one counts the chord lengths on the thin section assumed to be representative of the core. Figure 4.23 shows the pore volume distribution for Coconino sandstone. Thus, generally speaking, there are two types of morphologies for sandstones – Euclidean and fractal – and the pore volume of the rock may include any amount of porosity from the two types of morphology. There is almost no sedimentary rock that does not have any fractal component in its morphology. The fractality is the result of diagenetic processes that result in the deposition of clays on the grains' that make it rough. Krohn (1988b) measured the fractal properties of carbonate rocks and shales, and qualitatively found the same behavior as that of sandstones.

4.17.3

The Correlation Function Method

Measuring fractal properties of a given sample in terms of the correlation functions is perhaps the most unambiguous method of estimating the fractal dimension. In

this method, one measures the density–density autocorrelation function $C(r)$ at a distance $r = |r|$, defined by

$$C(r) = \frac{1}{V} \sum_{r'} s(r') s(r + r'), \quad (4.105)$$

where V is the volume of the sample. The point $r = 0$ is in the pore space and $s(r) = 1$ if a given point at a distance r from the origin is in the pore space, and $s(r) = 0$ otherwise. For a d -dimensional, self-similar system and large values of r , we must have

$$C(r) \sim r^{D_f - d} \quad (4.106)$$

since only a power law preserves the self-similarity. Therefore, D_f is estimated from a logarithmic plot of $C(r)$ versus r . Fara and Scheidegger (1961) were the first to use such statistical properties for characterizing porous media. In their method, one draws an arbitrary line through a porous medium. Points on this line are defined by giving them an arc length κ from an arbitrarily-selected origin. Certain values of κ correspond to the pore space, while others represent the solid matrix. A function $f(\kappa)$ is then defined such that $f = 1$ if the line at κ passes through the pore space, and $f = -1$, if the line at κ passes through the matrix. It is not difficult to show that $\langle f \rangle = 2\phi - 1$; $\langle f^n \rangle = \langle f \rangle$ if n is odd, and $\langle f^n \rangle = 1$ if n is even. A spectral analysis of f also provides information about the structure of the porous medium. The same basic idea was later used by others for obtaining the fractal properties of porous media (see below).

Images of a porous medium are used for computing $C(r)$ (Berryman and Blair, 1986). Samples of the porous medium are saturated with a low-viscosity epoxy, petrographic thin sections are prepared and polished, and the SEM in backscatter mode is used for producing high contrast images of the pore space and the solid matrix with several magnification. They are then digitized and stored on arrays of given sizes. The arrays are processed using digital image techniques to produce image of zeros and ones that closely approximate the matrix and pore space, which is then used for computing the various correlation functions. More advanced techniques, for example, X-ray computed tomography, may also be used for developing the images of a porous medium, although they require intensive computations and large computer memory on the order of gigabytes.

Katz and Thompson (1985) used an optical technique to measure the correlation functions using micrographs of polished thin sections that had been photographically enhanced to produce a binary image. Two identical negatives were made and placed in an optical microscope to measure the transmitted light through both films. The correlation function $C(r)$ was calculated as the transmitted intensity as a function of the distance that one film was translated relative to the other. Figure 4.24 shows the results for the Pico River sandstone in Utah (Thompson *et al.*, 1987a). The plot was made in the log–log scale because the deviations from a straight line provide estimates of the lower and upper limits, $l_{\min}$ and $l_{\max}$, of fractal behavior. The porosity of the sample sandstone was very low and had been highly altered by the diagenetic processes, so much so that the original sedimentary sandstone grains were difficult to recognize. If the alterations by the diagenetic process

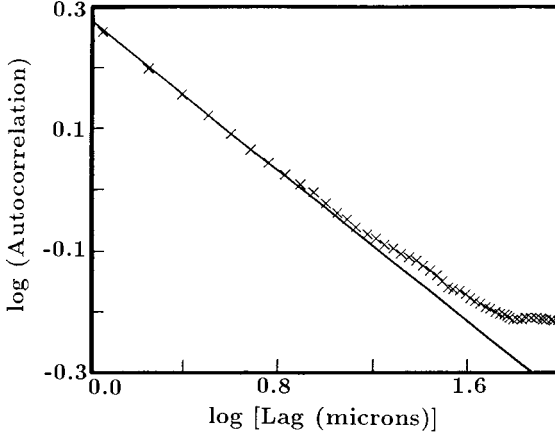


Figure 4.24 Autocorrelation function for Pico River sandstone (Utah), indicating the upper limit of fractality (after Thompson *et al.*, 1987a).

are not severe, then the pore space (volume) may not be fractal, and only the pore surface may have fractal properties. In such cases, the correlation function has a complex structure, even as a log-log plot. In some cases, for example, the Coconino sandstone, the pore space is fractal, but anisotropy gives rise to complex $C(r)$, even as a log-log plot. Another complicating factor is the presence of pores that are not connected, or are separated by more than a distance $l_{\max}$, implying that they are uncorrelated. In such cases, even the appearance of straight lines on log-log plots of $C(r)$ is *not* unambiguous evidence for fractal behavior of the pore space. Thus, although methods that use thin sections of porous media yield important information about their structure, they also have their limitations.

Berryman and Blair (1986) investigated the statistical properties of the function $s(\mathbf{r})$ used in Eq. (4.105) by constructing higher-order correlation functions (described in detail by Torquato, 2002 and Sahimi, 2003a; see also Chapter 7). If we define the following quantities

$$S_1 = \langle s(\mathbf{r}) \rangle, \quad (4.107)$$

$$S_2(\mathbf{r}_1, \mathbf{r}_2) = \langle s(\mathbf{r} + \mathbf{r}_1)s(\mathbf{r} + \mathbf{r}_2) \rangle, \quad (4.108)$$

$$S_3(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3) = \langle s(\mathbf{r} + \mathbf{r}_1)s(\mathbf{r} + \mathbf{r}_2)s(\mathbf{r} + \mathbf{r}_3) \rangle, \quad (4.109)$$

then, because two points lie along a line and three points in a plane, the statistics S_i may be measured by using images of cross sections of a porous medium. If one assumes that the porous medium is macroscopically homogeneous and isotropic, then it is not difficult to show that $S_2(\mathbf{r}_1, \mathbf{r}_2) = S_2(\mathbf{r}_2 - \mathbf{r}_1) = S_2(|\mathbf{r}_2 - \mathbf{r}_1|)$. Moreover,

$$S_1 = S_2(0) = \phi, \quad (4.110)$$

$$\lim_{r \rightarrow \infty} S_2(r) = \phi^2, \quad (4.111)$$

$$S_2'(0) = -\frac{1}{4}\Xi, \quad (4.112)$$

where Ξ is the specific surface area. Equation (4.112) was first derived by Debye *et al.* (1957).

4.17.4

Small-Angle Scattering

Small-angle scattering provides a measure of fractal behavior at length scales between 0.5 and 50 nm. In a scattering experiment, the observed scattering density $I(q)$ is given by the Fourier transform of the correlation function $C(r)$,

$$I(q) = \int_0^\infty C(r) \exp(i\mathbf{q} \cdot \mathbf{r}) d^3r, \quad (4.113)$$

where $\mathbf{q}$ is the scattering vector, the magnitude of which is given by Eq. (4.77). In a scattering experiment, $C(r)$ refers to the spatial variations in scattering amplitude per unit volume, rather than its physical density. It is not unreasonable to assume that in a porous medium with sufficiently low porosity, there is (to a good approximation) no interference scattering and, therefore, the total scattering intensity is the sum of the scattering from all the pores. For an isotropic medium, $C(\mathbf{r}) = C(r)$, where $r = |\mathbf{r}|$. Since the correlation function for a 3D fractal sample is given by Eq. (4.106) with $d = 3$, its substitution into Eq. (4.113) yields

$$I(q) \sim q^{-D_f} \Gamma(D_f - 1) \sin \left[\frac{(D_f - 1)\pi}{2} \right], \quad (4.114)$$

where Γ is the gamma function. Both light scattering and small-angle X-ray scattering from silica aggregation clusters confirmed the validity of Eq. (4.114) (Schaefer *et al.*, 1984).

In real porous media and materials, the range of scale-invariance and fractal behavior may be limited by lower and upper cutoffs $l_{\min}$ and $l_{\max}$. Finite size of a sample can also limit the fractal behavior. Under such conditions, the assumption of scattering by individual pores may break down and lead to interference scattering. To take into account the effect of a cutoff in the fractal behavior, Sinha *et al.* (1984) introduced into $C(r)$ an exponentially decaying term, incorporating a scattering correlation length ξ_s that reflects the upper limit of fractality, namely,

$$C(r) \sim r^{D_f-3} \exp \left(-\frac{r}{\xi_s} \right), \quad (4.115)$$

which, when used in Eq. (4.111), yields

$$I(q) \sim q^{-1} \Gamma(D_f - 1) \xi_s^{D_f-1} \sin \left[(D_f - 1) \tan^{-1}(q \xi_s) \right] \left[1 + (q \xi_s)^2 \right]^{\frac{(1-D_f)}{2}}. \quad (4.116)$$

The validity of Eq. (4.116) was confirmed by Sinha *et al.* (1984) for silica particle aggregates. Note that in the limit $\xi_s \rightarrow \infty$, we recover Eq. (4.114). However, for small values of $q \xi_s$ and $D_f = 3$ (homogeneous media), we obtain

$$I(q) \sim 8\pi \xi_s^2 \left[1 + (q \xi_s)^2 \right]^{-1}, \quad (4.117)$$

which is the classical result of Debye *et al.* (1957).

If r is small, that is, scattering at large q but small enough to be within the small-angle approximation, then the scattering reflects the nature of the boundaries between the pores and their surfaces, and then the scattering technique may be used to estimate the surface fractal dimension D_s , which may or may not be the same as the fractal dimension D_f of the pore space itself. Bale and Schmidt (1984) showed that for rough surfaces described by a fractal dimension $D_s > 2$, the correlation function takes on the following form:

$$C(r) \sim 1 - ar^{3-D_s}, \quad (4.118)$$

in which $a = A_0[4\phi(1-\phi)V]^{-1}$ and A_0 is a constant with the dimensions of area, and equal to the pore surface area when $D_s = 2$. Substituting Eq. (4.118) into Eq. (4.113) yields

$$I(q) \sim q^{D_s-6} \Gamma(5-D_s) \sin \left[\frac{(D_s-1)\pi}{2} \right], \quad (4.119)$$

which reduces to $I(q) \sim q^{-4}$, the classical result of Porod (1951) for smooth surfaces, $D_s = 2$, which is valid at the *shortest* length scales. Bale and Schmidt (1984) were able to confirm the validity of Eq. (4.119) for pores in lignites and sub-bituminous coals using SAXS (see Figure 4.25). If both the pore space and pore surface are fractal and, $D_f \neq D_s$, it is not difficult to show that

$$I(q) \sim q^{D_s-2D_f}. \quad (4.120)$$

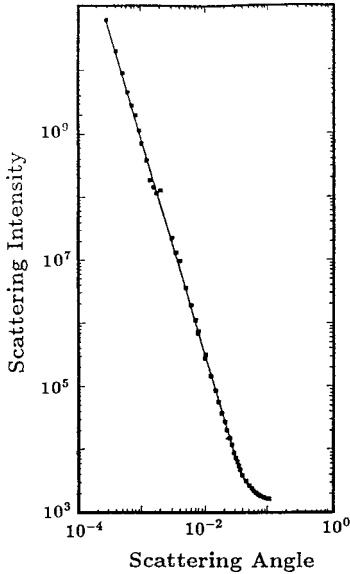


Figure 4.25 The scattering intensity for lignite coal. The scattering angle is in radians (after Bale and Schmidt, 1984).

Therefore, one has a crossover from the q^{-D_f} -dependence to the $q^{D_s-2D_f}$ and the q^{D_s-6} -dependence. The crossover between q^{-D_f} and q^{D_s-6} occurs at a scattering correlation length ξ_s such that $q \sim \xi_s^{-1}$. If $D_s \approx 3$, which is the case for some shaly rock (Mildner *et al.*, 1986), the crossover between the q^{-D_f} and q^{D_s-6} regimes may be difficult to discern.

Wong *et al.* (1986) used SANS and studied 26 different porous media, 12 of which were sandstone, 4 shales, 4 limestones, and 6 dolomites. Of the 16 sandstone and shale samples, 15 had a *fractal pore surface*, though *not* a fractal pore volume with $2.25 \leq D_s \leq 2.9$. The largest D_s was for a Coconino sandstone, consistent with Krohn (1988a)'s estimate. The lowest D_s was for Fontainebleau sandstone. The SEM images of Coconino sandstone indicated that the quartz grains are covered by clay, resulting in a convoluted surface and a large D_s , the SEM images of Fontainebleau sandstone showed that the quartz grains were very clean. Wong *et al.* (1986) also found that the carbonate rock they studied had very different properties than their sandstone and shaly rock samples, and was quite "clean", showing almost no trace of clays and, therefore, no diagenetic alteration.

Lucido *et al.* (1988) used the SANS on 18 volcanic rock samples and concluded that (1) the pore volumes of the rock samples were *not* fractal, and (2) it is not possible to determine from their data whether the pore surfaces were fractal. Hansen and Skjeltorp (1988) studied sandstone samples from 0.5–200 μm , and found that $D_f \simeq 2.7 \pm 0.05$, and $D_s \simeq 2.56 \pm 0.07$, which, to within the estimated errors of the experiments, are almost consistent with $D_f = D_s$.

Why should natural porous media (and fractured rock to be described in Chapter 6) have fractal properties? This is not completely understood yet, but there is little doubt, if any, that diagenetic processes play an important role in the formation of fractal rock. What is important to remember is that any realistic modeling of fluid transport and displacement processes in porous and fractured media must take into account the effect of such fractal properties.

4.17.5

Porosity and Pore Size Distribution of Fractal Porous Media

Katz and Thompson (1985) proposed that the porosity of fractal porous media can be estimated from

$$\phi = c \left(\frac{l_{\min}}{l_{\max}} \right)^{3-D_f}, \quad (4.121)$$

where c is a constant of order unity, and $l_{\min}$ and $l_{\max}$ are the lower and upper limits of fractal behavior. The predictions of Eq. (4.121) seem to agree well with the measured values, indicating the usefulness of D_f for estimating porosity of porous media. On the other hand, Pfeifer *et al.* (1984) proposed that in fractal porous media, the total volume V of pores with diameter $\geq 2r$ follows the following equation,

$$-\frac{dV}{dr} \sim r^{2-D_s}, \quad (4.122)$$

from which a pore size distribution may be deduced.

5

Characterization of Field-Scale Porous Media: Geostatistical Concepts and Self-Affine Distributions

Introduction

In Chapter 4, we described various techniques for characterization of laboratory-scale (LS) porous media and interpreting the data on their properties. However, although characterization of the LS porous media provides valuable information about and insight into their morphology, it is not by itself nearly enough for developing a realistic model of *field-scale* (FS) porous media, even if many coreplugs from such porous media are characterized. This is due to the fact that the FS porous media are heterogeneous over *multiple* and *distinct* length scales. However, characterization of the LS porous media provides information about the morphology over only the two smallest length scales, namely, the pore and the sample size, but not larger scales.

The characterization of a porous medium at length scales larger than the LS requires, in addition to data, use of special techniques and tools, and is collectively known as *geostatistics*. The purpose of this chapter is to describe the most important concepts and ideas of modern geostatistics. Advances in the measurement and estimation techniques, considerable progress in the development of theoretical and computational methods, and modern geostatistical techniques have made it possible to develop highly-resolved models of the FS porous media. Such resolved models, which usually contain a considerable amount of data on the spatial variations of the porosity, permeability, and other important properties, are known as the *geological models*. They typically consist of a three-dimensional (3D) (or sometimes 2D) computational grid with a few million grid blocks. The blocks' linear dimensions are on the order of several (or more) meters in the vertical direction and tens of meters areally, with their effective permeabilities and porosities distributed throughout according to the data and what the geostatistical analysis (see below) dictate. The grid blocks and their associated properties represent an intermediate scale between the LS and field scale.

However, due to computational limitations (mainly computation time), it is very difficult to carry out simulation of fluid flow and transport using the highly-resolved geological models. It is, therefore, necessary to *upscale* the properties of the grid blocks in the geological models in order to develop coarsened grids that not only can they be used in computer simulations with an affordable amount of compu-

tation time, but also yield accurate results. The grid blocks in the upscaled grids represent an intermediate step between the geological model and the field scale. Although, this is, in a sense, an artificial length scale introduced by computer simulators, it is a very important practical length scale. Many techniques for upscaling the geological models have been developed (see, for example, Rasaei and Sahimi, 2008, 2009a,b; Sahimi *et al.*, 2010).

To characterize and model the FS porous media, one must first analyze the available data. We divide the data into two groups:

1. In one group are what we call the *direct* data. Two important properties of the FS porous media, namely, the spatial distributions of their porosity and permeability, belong to this group. The spatial distribution of the porosity is measured along, for example, wells during drilling or estimated relatively accurately by indirect methods, such as, measuring the resistivity of many samples. The permeability distribution can either be estimated by in-situ nuclear magnetic resonance (NMR) (Mair *et al.*, 1999; see Chapter 9), or by coring and laboratory measurements.

Other types of logs, for example, the gamma ray and temperature logs, also belong to this class of data. If a porous medium is also fractured, the fractures' permeability is usually estimated by relating the fracture aperture to the fracture excess conductivity measured on electrical image logs (Sibbit, 1995), critically-stressed fractures within the present-day stress field, or both. However, such methods yield, at best, a relative estimate of the fracture permeability that must be calibrated against dynamic (flow) data. In any event, provided that enough amounts of the direct data are available, they are combined with the geostatistical methods (see below) to generate accurate spatial distributions of the porosity and permeability throughout a FS porous medium, and used in the generation of the geological model.

2. In the second group are what we call the *indirect* data, the most important of which are seismic recordings. Such data, when combined with a geostatistical method, provide information about the porosity distribution. If a FS porous medium is fractured, then, since the fractures are usually below the limit of seismic resolution, they cannot be directly detected by seismic experiments. As a result, the static models of fractures and fracture networks are mainly constrained by the direct data. Seismic data do, however, yield insight into the large-scale structure of geological formations, such as, their stratification and faults at the largest length scales. The static models include the mechanical origin (shear versus joint), the geometry (orientation, size, and frequency), and topology (open or connected versus isolated or cemented) of the fractures and their network. These aspects will be described in Chapter 6. As described there, seismic data can also be combined with the well-log data for generating more accurate models of a FS porous medium.

Sometimes, it may be very difficult to obtain meaningful direct data for some FS porous media. Dehghani *et al.* (1999) provided an excellent example of such a situ-

ation by describing their work for a carbonate porous formation, though the problem is general. The direct data are more useful if, in addition to being associated with specific depths and wells (hence, the region in which they are collected), they are also associated with a certain formation *type* or *facies*. In addition, despite years of study, identification of the spatial distribution of fractures and faults remains a largely unsolved problem. Until very recently, it was also believed that porosity *alone* cannot provide much insight into the whereabouts of the fractures. The permeability of fractures is much larger than that of the porous matrix in which they are embedded and, thus, data for the permeability provide insight into the spatial distribution of fractures. There is, however, an insufficient volume of the permeability data. Borehole-wall imaging is a very reliable method of mapping the intersections of the fractures and faults with the wells. Such imaging techniques are, however, expensive and are not always included in a logging run. In addition, they are not available for many of the older oil reservoirs and other FS porous formations.

Before starting the description and discussion of the various geostatistical concepts, let us point out that there are several excellent books on the principles of geostatistics, ranging from the pioneering books of Matheron (1967b) and Journel and Huijbregts (1978), to the more recent ones by Isaaks and Srivastava (1989), Cressie (1991), Wackernagel (1995), and Jensen *et al.* (2000). The book by Deutsch and Journel (1998) contains some very useful computer programs for use in practice. Thus, the discussions in the present chapter are not, and do not have to be, exhaustive. We provide a useful and understandable summary of the most important geostatistical concepts and techniques used in practice and invoked in the rest of this book. It is assumed that the reader is familiar with the basic concepts of the probability theory, although we recall the theory's necessary elements whenever necessary.

5.1

Estimators of a Population of Data

Given a set or population of data points, how does one evaluate the property of the porous formation for which the population was obtained? If we had the probability distribution function (PDF) of the population, then, using well-known laws of probability theory, we could compute any quantity that we wish. However, for any given population or sample with a limited number of data points, the task is to extract as much information and insight as possible from the population without necessarily knowing the PDF. This task is commonly referred to as *estimation*, and any method for computing a characteristic quantity for the data in the population, for example, the arithmetic or geometric average of the property for which the population is available, is called an *estimator*. As described by Jensen *et al.* (2000), estimation involves four essential ingredients that are as follows.

1. The characterizing statistic e to be evaluated, such as, various averages or the standard deviation.

2. The estimator $\mathcal{E}$.
3. The estimate $\hat{e}$.
4. The standard error or confidence interval of $\hat{e}$ that enables one to evaluate the accuracy of $\hat{e}$. The standard error is $\sigma(\hat{e})$, where σ^2 is the variance.

How many parameters should one estimate? The number depends on the PDF and what one intends to do with the statistics. For example, a Gaussian distribution is characterized by two parameters, its mean and the variance. The next question is, what are the properties of a good estimator? In general, an accurate estimator must have five properties: (1) small bias; (2) good efficiency; (3) robustness; (4) consistency, and (5) the ability to yield physically-meaningful estimates. An estimator's bias b is defined by

$$b = E(\hat{e}) - e, \quad (5.1)$$

where E denotes the expected value, or the expectation, of $\hat{e}$.¹⁾ In an ideal situation, $b = 0$, but, as usual, any practical situation is far from ideal and, therefore, $b \neq 0$. The two sources that give rise to a nonzero b are the precision of the measurements and the sampling.

The efficiency of an estimator is measured in terms of the variability of the estimates that it provides. If the estimator's variance is small, it is called an *efficient* or a *precise* estimator. Note that the sample's variance is *not* the same as the estimator's. Thus, an efficient estimator has a small $\sigma(\hat{e})$. It is very common (Jensen *et al.*, 2000) to have $\sigma^2(\hat{e}) \propto N^{-1}$, where N is the number of data points. At the same time, $\sigma^2(\hat{e}) \propto e^2$ is quite reasonable.

The robustness of an estimator is measured in terms of its insensitivity to errors in a small proportion of the data set. Hoaglin *et al.* (1983) and Barnett and Lewis (1984) provide detailed discussions of how to develop quantitative measures of robustness. The so-called *L-moments* (Hosking and Wallis, 1997; Jensen *et al.*, 2000) are used to measure the robustness of estimators. We define the moments

$$M_n = \int_0^1 x F^n dF, \quad (5.2)$$

where F is the cumulative distribution function of the stochastic variable X (i.e., the integral of its PDF). Then, the first four *L-moments* are given by $L_1 = M_0$ (which is just the mean value of X); $L_2 = 2M_1 - M_0$ (which is similar to the variance, except that it attributes smaller weights to the extreme values of X); $L_3 = 6M_2 - 6M_1 + M_0$ (which is a measure of the skewness of the distribution), and $L_4 = 20M_3 - 30M_2 + 12M_1 - M_0$ (which is a measure of peakedness of the distribution). Note that the integration in Eq. (5.2) is from zero to one because it is over all the probabilities, rather than all values of x . Because such moments attribute smaller weights to the extreme values than the usual moments of the PDF, they can be accurately

1) Recall that if $f(x)$ is the PDF of the random variable X , $E(X)$ is defined by, $E(X) = \int_{-\infty}^{\infty} x f(x) dx$, while for a discrete variable X , $E(X) = \sum_{i=1}^N p_i X_i$, where p_i is the probability that $X = X_i$.

estimated using much smaller data bases than would otherwise be needed, which is why they are valuable.

In general, quantitative assessment of an estimator robustness is difficult. If the errors in the data are small, and if we have an analytical expression for the estimator $\mathcal{E}$, then, based on the relation, $\hat{e} = \mathcal{E}(X_1, X_2, \dots, X_N)$, the sensitivities are calculated using a truncated Taylor series of $\mathcal{E}$ about the true values X_i ; that is, $\Delta \hat{e}_i = (\partial \mathcal{E} / \partial X_i) \Delta X_i$, where ΔX_i is the error in the measurement of the i th datum, assuming (implicitly) that the errors ΔX_i are independent. The total error is then the sum of the individual errors, $\sum_i \Delta \hat{e}_i$. A similar analysis can be done based on the variances, $\sigma^2(X_i)$.

If the arithmetic average $\bar{X}$ of a sample is the estimator, then its properties can be analyzed. According to the central-limit theorem, for large N , the size of the sample or population (number of the data points) $\bar{X}$ is normally distributed with the mean and variance of X_i , *regardless* of the PDF of X . If X is itself normally distributed, then $\bar{X}$ is *exactly* normally distributed. The sample median, $\bar{X}_{0.5}$, can be another estimator that is more robust than the sample mean $\bar{X}$, but for which the central-limit theorem cannot be used for obtaining its distribution.

Note that the variability of $\bar{X}$ decreases as N , the number of data points, increases. Also, note that if the X_i are distributed according to skewed distributions, such as the log-normal distribution which give larger weights to the very small or very large values, then the sample mean is inefficient and inaccurate. For example, since in many cases it is assumed that the permeabilities in a FS porous medium are distributed according to a log-normal distribution, their arithmetic average is a poor estimate of the effective permeability of the porous formation.

Now, consider the sample variance s^2 as the estimator (we use s^2 to distinguish it from σ^2 , the variance of X). If X is distributed according to a Gaussian distribution with mean μ and variance σ^2 , then, $\sigma_s^2(\hat{s}^2) = \sigma^2/N$, where $\hat{s}^2 = s^2/(N-1)$. More generally, regardless of the PDF of X , $\sigma_s^2(\hat{s}^2) = \{E[(X-\mu)^4] - \sigma^2(X)\}/[4(N-1)\sigma^2]$.

5.2

Heterogeneity of a Field-Scale Porous Medium

Thus far in this book, we have repeatedly mentioned heterogeneous porous media, by which we have meant variations in the shapes, sizes, and connectivity of the pores. However, this type of heterogeneity is more suitable for describing the disorder in the LS porous media. In case of the FS porous media, the heterogeneity is usually meant to be the *spatial variations* in the formation properties that affect *fluid flow* at large length scales. The permeability variation is, of course, one key factor that affects fluid flow and, therefore, the heterogeneity of the FS porous media may be defined in terms of such variations. In general, however, one may define a heterogeneity index in a variety of ways based either on a *static* property of a FS porous medium or a *dynamic* one. Detailed discussions of both classes of heterogeneity are presented by, among others, Jensen *et al.* (2000).

Static measures of the heterogeneity are based on the data taken from samples of porous media. Then, a model of fluid flow in the samples helps quantifying the effect on fluid flow of the variations (heterogeneity) in the property values. There are at least four static measures of the heterogeneity, and what follows is a brief description of each.

5.2.1

The Dykstra–Parsons Heterogeneity Index

The index proposed by Dykstra and Parsons Dykstra and Parsons (1950) is perhaps the most widely used measure of heterogeneity, and is defined by

$$H_{DP} = 1 - \frac{K_{0.16}}{K_{0.5}}. \quad (5.3)$$

Here, $K_{0.5}$ is the median permeability, while $K_{0.16}$ is the permeability at one standard deviation below $K_{0.5}$ in a log-probability plot. That is, $K_{0.5}$ and $K_{0.16}$ are the permeabilities if, in a plot of $\log K$ versus the probability scale, the probabilities are 0.5 and 0.16. For a completely uniform porous medium, $H_{DP} = 0$, whereas for an “infinitely” heterogeneous porous formation, $H_{DP} = 1$. The latter type of porous formation is a hypothetical one with one layer of infinite permeability and nonzero thickness.

Given a set of permeability data, one orders them in increasing values. With each data point is associated a probability that represents the length of the interval represented by the point. Although it was originally suggested that $K_{0.5}$ and $K_{0.16}$ should be obtained from the straight line that represents the best fit of the data in a log-probability plot, Jensen and Currie (1990) suggested to use the data directly. If $\ln K$ is normally distributed with a standard deviations σ_K , then Eq. (5.3) yields

$$H_{DP} = 1 - \exp(-\sigma_K). \quad (5.4)$$

In addition, it is important to recognize that H_{DP} (or any other heterogeneity index computed based on the permeability) varies significantly when it is computed based on vertical well data or areally, both of which are distributed normally, even if the permeabilities themselves are not.

If H_{DP} is estimated from vertical wells, one finds that $0.65 \leq H_{DP} \leq 0.99$, but if it is estimated areally from arithmetically-averaged well permeabilities, one has $0.12 \leq H_{DP} \leq 0.93$ (Lambert, 1981). Both estimates are distributed according to a Gaussian distribution, although most of the permeabilities are not distributed according to such a distribution. If one assumes that $\ln K$ is distributed according to a normal distribution, then the bias and standard error of H_{DP} can be derived analytically (Jensen and Lake, 1988):

$$b_{DP} = -0.749 \frac{[\ln(1 - H_{DP})]^2 (1 - H_{DP})}{N}, \quad (5.5)$$

$$\sigma_{DP} = -1.49 \frac{(1 - H_{DP}) \ln(1 - H_{DP})}{\sqrt{N}}. \quad (5.6)$$

Note that $b_{DP} < 0$, which means that the Dykstra–Parsons index *always underestimates* the heterogeneity, but the bias is typically small. Moreover, $b_{DP} \propto 1/N$ and attains its maximum for $H_{DP} \simeq 0.87$.

However, H_{DP} does have several weaknesses and, therefore, cannot be considered as a universal measure of the heterogeneity. Lambert (1981) provided evidence that H_{DP} cannot differentiate between formation types. When oil recovery from laboratory-scale waterfloods was correlated with H_{DP} (Lake and Jensen, 1991), it was observed that the models are not sensitive to the heterogeneity if $H_{DP} < 0.5$, but for broadly-distributed heterogeneities, the sensitivity was significant.

5.2.2

The Lorenz Heterogeneity Index

A heterogeneity index more accurate than the DP index is the so-called Lorenz index H_L , which is computed based on the data for the porosity *and* permeability. In this method, the data are first ordered according to decreasing values of K/ϕ . One then computes the fraction of the total flow capacity defined by

$$F_M = \frac{\sum_{m=1}^M K_m h_m}{\sum_{n=1}^N K_n h_n}, \quad (5.7)$$

and the fraction of total storage capacity defined by

$$C_M = \frac{\sum_{m=1}^M \phi_m h_m}{\sum_{n=1}^N \phi_n h_n}, \quad (5.8)$$

where N is the number of data points, h_m the thickness of the layer m in which the porosity and permeability are ϕ_m and K_m , and $1 \leq M \leq N$. A plot of F_M versus C_M is then prepared (which should pass through the origin and the point (1, 1)). The area A between the curve and the diagonal straight line [passing through (0,0) and (1,1)] is then computed numerically. The Lorenz heterogeneity index H_L is then given by $H_L = 2A$.

Similar to H_{DP} , the Lorenz index is also zero for completely homogeneous porous media and approaches one for infinitely heterogeneous porous formations. Typical values of H_L for field-measured values of the permeability and porosity are $0.6 \leq H_L \leq 0.9$. If $\ln K$ is distributed according to a Gaussian distribution with a standard deviation σ_K , then the Lorenz and DP indices are related:

$$H_L = \exp\left(\frac{1}{2}\sigma_K\right) = \operatorname{erf}\left[-\frac{1}{2}\ln(1 - H_{DP})\right], \quad (5.9)$$

where erf denotes the error function. In general, it can be shown that the errors in the estimates of H_L are smaller than those in H_{DP} .

Although H_L is somewhat more difficult to compute than H_{DP} , it does have several advantages over H_{DP} that are as follows (Jensen *et al.*, 2000).

1. It can be estimated accurately for any distribution.
2. It does not rely on the best fits of the data, as does H_{DP} .
3. It depends on both the porosity ϕ and layer thickness h .
4. If a porous medium consists of N uniformly stratified elements between wells through which a fluid is flowing, then F_M represents the fraction of the total flow passing through a fraction C_M of the medium's volume. Therefore, in this particular case, the Lorenz coefficient has a clear physical interpretation. Other useful features of H_L are described by Jensen *et al.* (2000).

The bias b_L in estimating the true value of H_L can be quite pronounced, particularly if H_L is large and the number of data points is relatively small. Similar to H_{DP} , the Lorenz index *underestimates* the heterogeneity. The standard error of H_L is also usually smaller than that of H_{DP} .

5.2.3

The Index of Variation

Another popular measure of the heterogeneity is the so-called *index of variation*, H_v , defined by

$$H_v = \frac{\sigma_K}{E(K)}, \quad (5.10)$$

where $E(K)$ is the expected value of the permeability. In most cases, the mean and standard deviations of the permeability data from different sources or populations vary in the same direction so that H_v remains fairly constant. Hence, if two sample data or populations are characterized by two very different values of H_v , the implication is that there must be fundamental structural differences between the two samples.

As discussed by several authors (see, for example, Hald, 1952; Koopmans *et al.*, 1964; Jensen *et al.*, 2000), it is difficult to analyze the statistical properties of the estimators of H_v . However, if the permeability is distributed according to a log-normal distribution, then an estimate of H_v can be derived. Suppose that K_A and K_H represent, respectively, the arithmetic and harmonic averages of the permeability data. Then, an estimate of H_v is given by (Johnson and Kotz, 1970)

$$\hat{H}_v = \left(\frac{K_A}{K_H} - 1 \right)^{\frac{1}{2}}. \quad (5.11)$$

The effect of H_v on unstable miscible displacements has been extensively studied, usually based on the assumption that H_v is a good measure of the heterogeneity. H_v is also useful when one compares variabilities of different facies (Jensen *et*

al., 2000). Studies by Kittridge *et al.* (1990) and Goggin *et al.* (1992) indicated that if one compares geologically-similar elements in outcrop and subsurface samples, the value of H_v remains largely unchanged even though there are large changes in the average permeabilities of the two distinct samples.

5.2.4

The Gelhar–Axness Heterogeneity Index

Gelhar and Axness (1983) introduced a heterogeneity index defined by

$$H_{GA} = \sigma_{\ln K}^2 \lambda_D, \quad (5.12)$$

where $\sigma_{\ln K}^2$ is the variance of the distribution of $\ln K$, and λ_D is the autocorrelation length in the macroscopic direction of fluid flow. Physically, λ_D represents a length scale over which similar values of the permeability exist (see also below). In other words, λ_D is a correlation length for positive correlation between the permeability values. It is precisely due to λ_D that the Gelhar–Axness index is, in many ways, superior to the three heterogeneity indices already defined. That is, whereas H_{DP} , H_L and H_v ignore such correlations, H_{GA} does not. Indeed, studies by Kempers (1990) and Waggoner *et al.* (1992) suggested that H_{GA} is a much better indicator of flow performance – and the effect of the heterogeneity on it – than the widely used H_{DP} ; see also the discussion by Sorbie *et al.* (1994).

5.2.5

The Koval Heterogeneity Index

The Koval index H_K (Koval, 1963) is an empirical way of quantifying the effect of the heterogeneity on miscible displacements. It is a dynamic index and is defined by

$$H_K = t_{BT}^{-1}, \quad (5.13)$$

where t_{BT} is the dimensionless time for the breakthrough – that is, when the displacing fluid first arrives at the outlet – in a miscible displacement in which the mobility ratio is one (see Chapter 13 for the description of miscible displacements). The definition of H_K also implies that the maximum oil recovery is obtained when H_K pore volumes of the displacing fluid has been injected. Thus, large values of H_K are detrimental to an efficient miscible displacement. The advantage of H_K over H_{DP} and H_L is that it is a more linear measure of the performance of a miscible displacement.

5.3

Correlation Functions

The next important subject is the correlations between various properties of a FS porous medium. Many of such correlations are empirical, as there are no rigorous

theoretical foundations that relate one property, say the permeability, to another property such as the porosity. Thus, although important, we do not consider them in this chapter.

However, in order to generate a realistic model of a FS porous medium, one must understand *autocorrelation*, that is, the correlation of a variable with *itself*. To better understand this point, suppose that we are given the porosity logs along several widely-separated wells in an oil reservoir. To develop a model of the reservoir, we must be able to estimate the porosity in the interwell zones for which there are no data. If the porosities were totally random (no correlation between them), then there would be no hope for developing any accurate model of the reservoir. The porosities are not, however, random, but are in fact highly correlated. This type of correlation is what is usually referred to as the autocorrelation, that is, the correlation between properties of the same sample space (in this example, the oil reservoir), *but at different locations* (between the porosities along the wells and those in the interwell zones). Understanding such autocorrelations and being able to construct accurate models for them is an important task in any study of a FS porous medium.

5.3.1

Autocovariance

Suppose that $Z(x)$ represents a property measured at x . In practice, the measurements are done at discrete points $x_1, x_2, \dots$, so that $Z_i = Z(x_i)$ represents the measured property at x_i . The autocovariance $R(Z_i, Z_j)$ between data Z_i and Z_j is defined by

$$R(Z_i, Z_j) = E \{ [Z_i - E(Z_i)][Z_j - E(Z_j)] \} = E(Z_i Z_j) - E(Z_i)E(Z_j), \quad (5.14)$$

where, as usual, $E(x)$ denotes the expected value of x . $R(Z_i, Z_j)$ describes the correlation between Z_i and Z_j as well as the effect of the heterogeneity. In practice, it is difficult to separate the two effects. Note that, $R(Z_i, Z_j) = R(Z_j, Z_i)$ and $R(Z_i, Z_i) = \sigma^2(Z_i)$, where σ^2 is the variance. Given a set of data, an estimate $\hat{R}$ of the true autocovariance is given by

$$\hat{R}(Z_i, Z_{i+m}) = \frac{1}{N-m} \sum_{i=1}^{N-m} (Z_i - \bar{Z})(Z_{i+m} - \bar{Z}), \quad (5.15)$$

where $\bar{Z}$ is an estimate for the mean value of the variable Z .

5.3.2

Autocorrelation

The autocorrelation coefficient $C_R(Z_i, Z_j)$ is defined by

$$C_R(Z_i, Z_j) = \frac{R(Z_i, Z_j)}{\sigma(Z_i)\sigma(Z_j)}. \quad (5.16)$$

Clearly, $-1 \leq C_R \leq +1$. The *extent* of the autocorrelation is sometimes quantified by an *integral scale* ξ_I defined by

$$\xi_I = \int_0^{\infty} C_R(r) dr, \quad (5.17)$$

which is defined for any autocorrelation with a finite correlation length. Note that if the random variable Z_i is independent, then, the variance $\sigma^2(\bar{Z})$ of the sample mean is simply

$$\sigma^2(\bar{Z}) = \frac{\sigma^2(Z_i)}{N}. \quad (5.18)$$

If, however, the random variable is dependent, then

$$\sigma^2(\bar{Z}) = \frac{1}{N^2} \sum_{i=1}^N \sum_{j=1}^N R(Z_i, Z_j) = \frac{\sigma^2(Z_i)}{N} \left[1 + \frac{2}{N} \sum_{i=1}^N \sum_{j=1}^N C_R(Z_i, Z_j) \right]. \quad (5.19)$$

For detailed analysis and discussions of *cross-correlations* between various types of data see Dashtian *et al.* (2011b).

5.3.3

Semivariance and Semivariogram

The semivariance is defined by

$$\gamma(Z_i, Z_j) = \frac{1}{2} E \left[(Z_i - Z_j)^2 \right]. \quad (5.20)$$

Clearly, $\gamma \geq 0$. A consequence of the nonnegativity of γ is that a semivariance can be noncontinuous *only* at the origin. In that case, the height of the jump or discontinuity is usually called the *nugget*. If the autocovariance of a random function $Z(x)$ exists, it is related to the semivariance by

$$2\gamma(x, y) = R(x, x) + R(y, y) - 2R(x, y) + E[Z(x) - Z(y)]^2. \quad (5.21)$$

One may also consider a normalized semivariance:

$$\gamma_n(Z_i, Z_j) = \frac{\gamma(Z_i, Z_j)}{\sigma(Z_i)\sigma(Z_j)}. \quad (5.22)$$

An estimate $\hat{\gamma}$ for the true semivariance is usually obtained from

$$2\hat{\gamma} = \frac{1}{N-m} \sum_{i=1}^{N-m} (Z_i - Z_{i+m})^2. \quad (5.23)$$

One may establish simple relations between the various quantities defined so far if the data are stationary. A strictly stationary series is one for which all the

moments are independent of the position, that is, the moments are translationally invariant. Such a series is very rare. In practice, however, we usually require up to second-order stationarity, that is, translational invariance of the first two moments. Assuming that the data are second-order stationary, we have $R(Z_i, Z_{i+m}) = R(m\Delta h) = R(h)$, where Δh is the distance between two neighboring points in the series and $h = m\Delta h$ is the *lag*, or the *lag distance*. The second-order stationarity of the data also implies that the estimate $\hat{R}(h)$ of the true autocovariance is given by

$$\hat{R}(h) = \frac{1}{N-m} \sum_{i=1}^{N-m} Z_i Z_{i+m} - \bar{Z}^2. \quad (5.24)$$

Similarly, for second-order stationary data, we have $C_R(Z_i, Z_{i+m}) = C_R(h)$, and $\gamma(Z_i, Z_{i+m}) = \gamma(h)$. The plot of $\gamma(h)$ versus h is called the *semivariogram*, while the plot of $R(h)$ versus h is referred to as the *autovariogram*. More importantly, one has a simple relation:

$$\gamma(h) = \sigma^2 - R(h), \quad (5.25)$$

which holds so long as the variance σ^2 is finite. Moreover, $C_R(h)$ and $\gamma(h)$ follow opposite trends: If $C_R(h)$ decreases, then $\gamma(h)$ increases. Note that if a random field $Z(x)$ is stationary and ergodic (i.e., the volume average of the field is equal to its ensemble average), then

$$\lim_{h \rightarrow \infty} \gamma(h) = \sigma^2[Z(x)], \quad (5.26)$$

that is, the limit of the semivariogram is equal to the variance of the field. This limit is usually called the *sill*. The distance over which the difference between the semivariogram $\gamma(h)$ and its sill becomes negligible is called the *range*.

If the data contain no stochasticity (which, for any set of data for a disordered porous medium, happens rarely, if ever), then the semivariogram increases monotonically with increasing lags. Step changes (increases) and quasi-periodic variations are also sometimes obtained. If a porous medium is anisotropic, then one will have distinct semivariograms in the horizontal and vertical directions. The opposite is also true: distinct semivariograms in distinct directions imply anisotropy.

Characterization of the autocorrelations in natural porous media is not as clean-cut. As described by Jensen *et al.* (2000), autocorrelations in clastic formations are typically scale-dependent. Moreover, many natural porous formations are anisotropic, with the anisotropy caused by stratification. For such porous media, the horizontal and vertical autocorrelations may be completely distinct. The vertical autocorrelations in clastic porous formations are usually weak, but they are strong in the horizontal direction. One may even have anisotropy in a horizontal plane that is caused by a unidirectional deposition process, or by a unidirectional flow, as in fluvial porous formations. Characterizing the autocorrelations is more difficult if there exist multiple distinct and relevant length scales in the porous formation. The stratification may not be so clear in carbonate porous formations, but the anisotropy does usually exist.

5.4

Models of Semivariogram

Empirical variograms cannot be computed at every lag distance h , simply because there are rarely enough data to do so. Moreover, due to variations in the estimation, it is not even guaranteed that one obtains a valid semivariogram – valid in the sense defined above. On the other hand, some geostatistical methods that are used for generating models of the FS porous media, such as Kriging and co-Kriging methods (see below), require valid semivariograms. Thus, the experimental semivariograms are often approximated by models that guarantee the validity of the semivariogram.

5.4.1

The Exponential Model

The model is given by

$$\gamma(h) = (\sigma^2 - \sigma_0^2) \left[1 - \exp\left(-\frac{\alpha h}{\xi_e}\right) \right] + \sigma_0^2 \delta_{(0,\infty)}(h). \quad (5.27)$$

The parameter ξ_e is the range that, physically, represents an autocorrelation length. α is a purely numerical parameter. σ^2 is the sill of the semivariogram, while σ_0^2 is the nugget.

5.4.2

The Spherical Model

The spherical variogram is defined by

$$\gamma(h) = (\sigma^2 - \sigma_0^2) \left\{ \left[\frac{3}{2} \left(\frac{h}{\xi_s} \right) - \frac{1}{2} \left(\frac{h}{\xi_s} \right)^3 \right] \delta_{(0,\xi_s)}(h) + \delta_{(\xi_s,\infty)}(h) \right\} + \sigma_0^2 \delta_{(0,\infty)}(h). \quad (5.28)$$

The spherical model is used extensively in modeling of the FS porous media. The parameters σ^2 , σ_0^2 and ξ_s represent, respectively, the sill (which is approximately the sample variance), the nugget (which can be measured directly), and the range or the autocorrelation length. If the data indicate that nugget $\approx$ sill, the semivariogram is said to exhibit a *pure nugget effect*.

5.4.3

The Gaussian Model

The Gaussian model of a semivariogram is given by

$$\gamma(h) = (\sigma^2 - \sigma_0^2) \left[1 - \exp\left(-\frac{\alpha h^2}{\xi_g^2}\right) \right] \delta_{(0,\xi_g)}(h) + \sigma_0^2 \delta_{(\xi_g,\infty)}(h). \quad (5.29)$$

The physical meanings of σ^2 , σ_0^2 , and ξ_g are the same as before. In all the three models introduced so far, the function $\delta_{(x,y)}(h)$ is defined by

$$\delta_{(x,y)}(h) = \begin{cases} 1 & \text{if } h \in (x, y), \\ 0 & \text{otherwise.} \end{cases} \quad (5.30)$$

5.4.4

The Periodic Model

Some empirical semivariograms exhibit a degree of periodicity or quasi-periodicity. Therefore, it is important to have a model that can accurately describe such semivariograms. One such model is given by

$$\gamma(h) = \sigma_0^2 + (\sigma^2 - \sigma_0^2) \left[1 + \left(\frac{\xi_p}{h} \right) \sin \left(\frac{h}{\xi_p} \right) \right]. \quad (5.31)$$

The model contains peaks and valleys that are the solutions of the equation, $h/\xi_p = \tan(h/\xi_p)$, with the first peak and valley being at $h/\xi_p = 3\pi/2$ and $h/\xi_p = 5\pi/2$, respectively. In many sedimentary formations, deeper valleys in a particular direction are common, implying the need for an anisotropic semivariogram model. Then, one can use a model such as

$$\gamma(h) = \sigma_0^2 + (\sigma^2 - \sigma_0^2) \left[1 - \cos \left(\frac{h}{\xi_p} \right) \right], \quad (5.32)$$

in the direction in which the deeper valleys are seen in the semivariogram.

5.5

Infinite Correlation Length: Self-Affine Distributions

There is increasing evidence that many FS porous media do not have a finite correlation length, but an infinitely long one. While the concept of an infinite correlation length may seem abstract, it does have a clear physical interpretation: the correlation length is as large as the linear size of the porous formation. The larger the size, the longer is the correlation length.

To describe such geological formations, a nonstationary stochastic process called the *fractional Brownian motion* (FBM) has been used. We first consider the 1D case, and define the FBM process $B_H(x)$ by (Mandelbrot and van Ness, 1968)

$$B_H(x) - B_H(0) = \frac{1}{\Gamma(H + \frac{1}{2})} \left[\int_{-\infty}^x K(x-s) dB(s) \right], \quad (5.33)$$

where x can be a spatial or temporal variable, $\Gamma(x)$ is the gamma function, and H is called the *Hurst* exponent. As described in Chapter 6, the FBM is also used to

describe and model rough surfaces, in which case it is referred to as the *roughness* exponent. The kernel $K(x - s)$ is given by

$$K(x - s) = \begin{cases} (x - s)^{H-\frac{1}{2}} & 0 \leq s \leq t \\ (x - s)^{H-\frac{1}{2}} - (-s)^{H-\frac{1}{2}} & s < 0. \end{cases} \quad (5.34)$$

It is not difficult to show that

$$B_H(bx) - B_H(0) \equiv b^H [B_H(x) - B_H(0)], \quad (5.35)$$

where “ $\equiv$ ” means “statistically equivalent to.” Equation (5.35) indicates that the FBM defines a *self-affine* stochastic distribution (Feder, 1988). Self-affinity is similar to self-similarity described in Chapters 3 and 4, except that it is direction dependent. That is, the self-similarity of the object or the distribution in different directions is by different scale factors (see also Chapter 6). To demonstrate that the FBM generates stochastic series with an infinite correlation length (or time), consider the correlation function $C(x)$ of “future” increments $B_H(x)$ with the “past” increments $-B_H(-x)$, defined by

$$C(x) = \frac{\langle -B_H(-x) B_H(x) \rangle}{\langle B_H(x)^2 \rangle}. \quad (5.36)$$

It is straightforward to show that $C(x) = 2(2^{2H-1} - 1)$ independent of x ; that is, the correlations persist everywhere.

The type of the correlations can be tuned by varying the Hurst exponent H . If $H > 1/2$, then the FBM displays *persistence* or *positive correlations*, that is, a trend (for example, a high or a low value) at x is likely to be followed by a similar trend at $x + \Delta x$, whereas with $H < 1/2$, the FBM generates *antipersistence* or *negative correlations*, that is, a trend at x is not likely to be followed by a similar one at $x + \Delta x$. For $H = 1/2$, the past and future are not correlated and, thus, the increments in $B_H(x)$ are completely random and uncorrelated.

The 1D FBM may be generalized to 2D or 3D. Hence, if we consider two arbitrary points $\mathbf{x}$ and $\mathbf{x}_0$ in 2D or 3D space, the FBM is defined by

$$\langle [B_H(\mathbf{x}) - B_H(\mathbf{x}_0)]^2 \rangle \sim |\mathbf{x} - \mathbf{x}_0|^{2H}. \quad (5.37)$$

Figure 5.1 presents 1D profiles generated by the FBM. The FBM is not a stationary process, though its increments are, although they are not ergodic. The variance of a FBM for a large enough array is divergent, implying that the variance increases with the size of the series without bounds (see below). The FBM trace in d dimensions is a self-affine fractal with a *local* fractal dimension $D_f = d + 1 - H$. The FBM is not differentiable at any point, but by smoothing it over an interval one obtains its approximate numerical derivative which is called the *fractional Gaussian noise* (FGN). The semivariogram of a FBM is given by

$$\gamma(h) = \gamma_0 h^{2H}, \quad (5.38)$$

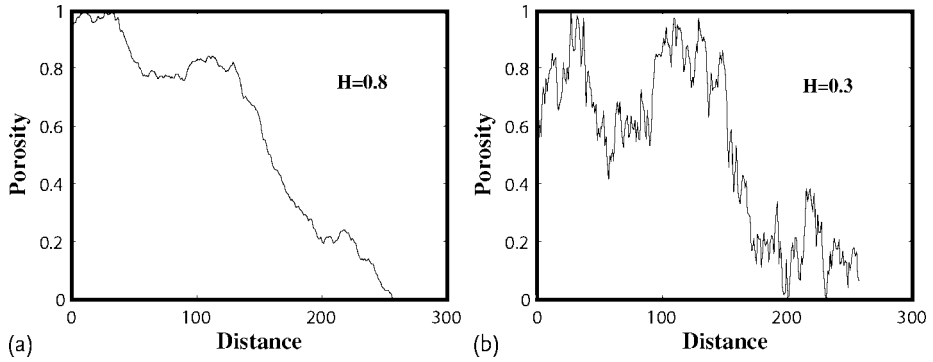


Figure 5.1 Examples of the 1D FBM: (a) $H = 0.8$; (b) $H = 0.3$ (courtesy of Dr. Fatemeh Ebrahimi).

where $\gamma_0 = \gamma(h = 1)$. Due to its simplicity and elegance, and the fact that the FBM does seem to describe accurately considerable amount of data (see below), the use of a FBM in the simulation of the FS porous media has become popular. The semivariogram of a 1D FGN is given by (Bruining *et al.*, 1997)

$$\gamma(h) = \gamma_0 s^{2H} - \frac{\gamma_1}{2s^2} [(h+s)^{2H} - 2h^{2H} + |h-s|^{2H}] , \quad (5.39)$$

where s is a smoothing parameter, and γ_0 and γ_1 are two constants.

In a pioneering work, Hewett (1986) presented evidence that the porosity logs of the FS porous media may follow the statistics of a FBM or FGN. More precisely, Hewett provided the first concrete evidence that the porosity logs in the direction perpendicular to the bedding may follow the statistics of a FGN, while those parallel to the bedding may follow a FBM. Since Hewett's original work, extensive studies by several research groups have provided compelling evidence for the validity of Hewett's proposal (Crane and Tubman, 1990; Sahimi and Yortsos, 1990; Taggart and Salisch, 1991; Aasum *et al.*, 1991; Hardy, 1992; Sahimi and Mehrabi, 1999; Sahimi *et al.*, 1995). For example, Crane and Tubman (1990) analyzed three horizontal wells and four vertical wells in a carbonate reservoir, and found that the porosity logs are described well by a FGN with $H \simeq 0.88$. Likewise, Hardy (1992) analyzed 240 porosity logs from a carbonate formation and found them to be described well by a FGN with $H \simeq 0.82$. The analysis of a single horizontal well in the same formation also indicated the FGN statistics with a slightly higher H . The analysis of porosity logs of a major Iranian oil reservoir (Sahimi and Mehrabi, 1999) indicated that at least some of the logs are described reasonably well by a FGN (most Iranian oil reservoirs are of carbonate-type). In all such studies, the lower limit of self-affine fractal behavior was *below* the resolution of the instruments used for measuring the data. A surprising discovery of most of the studies was that the same value of H was found for the porosity and resistivity logs that, in general, is not expected to be the case.

Similarly, permeability measurements on outcrop surfaces have provided additional evidence that the FBM- and FGN-type behavior in oil reservoirs and other

natural formations is not the exception, but the rule. For example, the analysis of Goggin *et al.* (1992) on sandstone outcrop data indicated that the logarithm of the permeabilities along the lateral trace followed the FGN statistics with $H \simeq 0.85$.

There is also evidence that some properties of oil reservoirs follow the statistics of a FBM. In the original analysis of Hewett (1986), a FBM was used for generating the horizontal properties of the reservoir. This was justified based on the data obtained from groundwater plumes (see, for example, Pickens and Grisak, 1981). Emanuel *et al.* (1989), Mathews *et al.* (1989), and Hewett and Behrens (1990) all obtained accurate descriptions of oil reservoirs using a FBM. Neuman (1994) provided further evidence that the permeability distributions of many aquifers follows the statistics of a FBM with $H < 0.5$. Makse *et al.* (1996b) analyzed data for two sandstone samples from distinct environments, and found the permeability distribution to be described by a FBM with $H \simeq 0.82\text{--}0.9$. Moreover, analyzing extensive data for the elastic moduli, the density, and seismic wave speeds in eight offshore and onshore oil and gas reservoirs, Sahimi and Tajar (2005) showed that even such properties follow the statistics of the FBM. Dashtian *et al.* (2011a) showed, in fact, that practically all type of well logs can be described by self-affine distributions.

Note that, as pointed out earlier, a FBM is a nonstationary stochastic process. However, if the self-affine fractal behavior indicated by the FBM is only manifested below a low-wave number cutoff $1/L$, then the permeability or porosity field is effectively stationary at length scales larger than L , with the variance given by

$$\sigma^2 = \frac{c}{2H} L^{2H}, \quad (5.40)$$

where c is a constant. Equation (5.40) demonstrates an earlier assertion that the variance of a FBM increases with the size L of the system without bound. A comprehensive discussion of such issues is given by Molz *et al.* (2004).

In general, the description of a FS porous medium by a FBM does not differ much from that by a FGN if the wells are closely spaced. Moreover, both the FBM and FGN descriptions approach a stratified medium as the variations in the porous medium properties between the wells decrease with decreasing well spacing. We should, however, point out that under practical circumstances, one encounters with the FS porous media, a FGN with $0 < H < 1$, producing results that are very similar to those obtained with the FBM with $1 < H < 2$, provided that one uses $H + 1$ instead of H in Eq. (5.38).

If the data contain long tails, then an FBM or FGN may not accurately describe them. For example, if an oil reservoir or aquifer is fractured, then one must detect large jumps in the permeability distribution, indicating the presence of the fractures since as a probe moves along a well and passes from the porous matrix to the fractures, the permeability should significantly increase. Compared with the number of pores of any geological formation, the fractures are relatively rare and, therefore, their presence represents rare events manifested by large jumps in the permeabilities and giving rise to long tails in their distribution that are not seen in data that follow the statistics of a FBM or FGN. In this case, a fractional Lévy motion (a generalization of a FBM) and the associated Lévy-stable distribution may

provide a more accurate and complete characterization of the data (Mehrabani *et al.*, 1997). We will come back to this point in the next chapter.

To give the reader some idea about the complexities that are involved in the log data, we show in Figure 5.2 a vertical porosity log that was collected along a vertical well in an oil reservoir in southern Iran (Sahimi and Mehrabi, 1999). The well's depth is about 1900 m and the porosity ϕ was estimated or measured every 20 cm so that over 600 data points were collected. The question then is: given a porosity log, or any other log or the permeabilities along the same well of a FS porous medium, how can one accurately analyze it to uncover its mathematical structure? In particular, if such data follow the statistics of a FBM or FGN, how can one estimate the Hurst exponent H that characterizes the correlations in the data?

A closely related issue is the efficient and accurate numerical simulation and generation of an array that follows the statistics of a FBM or a FGN if they describe the porosity logs, permeability distributions, and other properties of a FS porous medium. In particular, such numerical simulations are needed when one uses the conditional simulations described in Section 5.7. Efficient and accurate simulation of a FBM or FGN is not straightforward at all. There are several techniques for numerical simulation and generation of an FBM array with a given Hurst exponent H (Mehrabani *et al.*, 1997). For example, Hamzehpour and Sahimi (2006a) developed a method by which a FBM array is generated by an optimization method using simulated annealing (see Chapters 6–8). We now describe four efficient methods for generation of a FBM or FGN.

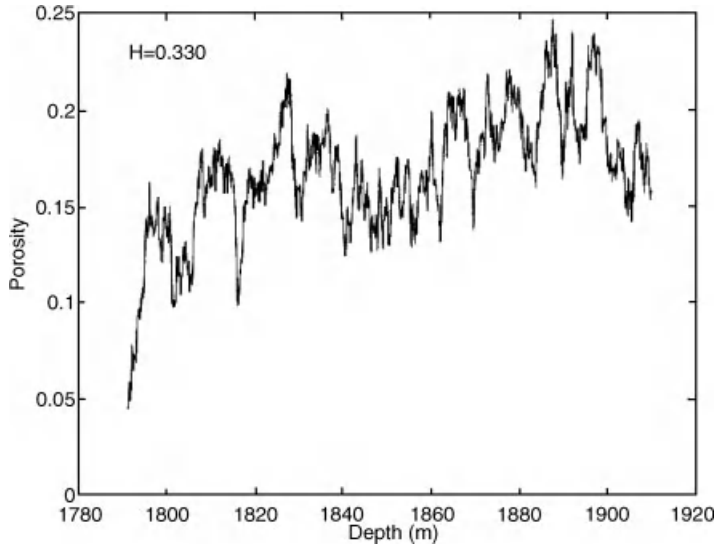


Figure 5.2 A porosity log along a well in a fractured oil reservoir (after Sahimi and Mehrabi, 1999).

5.5.1

The Spectral Density Method

A convenient way of representing a stochastic function is through its spectral density $S_d(\omega)$, the Fourier transform of its two-point correlation function in d -dimensions. Bruining *et al.* (1997) derived the spectral density of the FBM. The result for a 1D FBM is given by

$$S_1(\omega) = \frac{H\gamma_0}{\Gamma(1-2H)\cos(H\pi)} \frac{1}{\omega^{2H+1}}. \quad (5.41)$$

Equation (5.41) indicates why a FBM is not differentiable because in order to be so, its spectral density should decay faster than ω^{-3} , which is not the case even for $H = 1$. Using a formula due to Tsuji (1955),

$$S_2(\omega) = \frac{1}{\pi} \int_0^\infty \frac{d}{d\omega_1} \left[\frac{1}{\omega_1} \frac{d}{d\omega_1} S_1(\omega_1) \right] \sqrt{\omega_1^2 - \omega^2} d\omega_1, \quad (5.42)$$

where ω denotes the magnitude of ω , one obtains the corresponding 2D density:

$$S_2(\omega) = \frac{2^{2H}\gamma_0 H^2}{\pi^2} \frac{\Gamma(H)^2 \sin(H\pi)}{(\omega_x^2 + \omega_y^2)^{H+1}}. \quad (5.43)$$

Tsuji (1955) also showed that

$$S_3(\omega) = -\frac{1}{2\pi\omega} \left[\frac{dS_1(\omega_1)}{d\omega_1} \right]_{\omega_1=\omega}, \quad (5.44)$$

and, therefore,

$$S_3(\omega) = \frac{\gamma_0 H}{2\pi\Gamma(1-2H)\cos(H\pi)} \frac{2H+1}{(\omega_x^2 + \omega_y^2 + \omega_z^2)^{H+\frac{3}{2}}}. \quad (5.45)$$

Note that the spectral representation of a FBM also allows one to introduce a cutoff length scale $\ell_c = 1/\omega_c$ such that

$$S_d(\omega) = \frac{a_d}{(\omega_c^2 + \omega_x^2 + \omega_y^2 + \omega_z^2)^{H+\frac{d}{2}}}, \quad (5.46)$$

where a_d is a d -dependent constant. The cutoff ℓ_c allows one to control the length scale over which the spatial properties of a porous formation are correlated, or anti-correlated. Thus, for length scales $L < \ell_c$, the properties preserve their correlations or anticorrelations, whereas for $L > \ell_c$, they become random and uncorrelated.

The spectral density for a 1D FGN is given by

$$S_1(\omega) = \frac{2}{\pi s^2} \gamma_0 H \Gamma(2H) \sin(\pi H) [1 - \cos(s\omega)] \frac{1}{\omega^{2H+1}}. \quad (5.47)$$

In the limit $s \rightarrow 0$, Eq. (5.47) reduces to

$$S_1(\omega) = \frac{1}{\pi} \gamma_0 H \Gamma(2H) \sin(\pi H) \frac{1}{\omega^{2H-1}}. \quad (5.48)$$

The spectral density of an FBM or FGN provides a convenient method for their numerical generation using a fast Fourier transformation (FFT) technique. In this method, one first generates random numbers, distributed either uniformly in $[0, 1]$, or according to a Gaussian distribution with random phases, and assigns them to the sites of a d -dimensional lattice. In most cases, the linear size L of the lattice is a power of two, but the only requirement is that L can be partitioned into small prime numbers so that the FFT algorithm can be used. One must also keep in mind that since the variance of a FBM increases with the size L of the array or lattice, generating a FBM array with a given variance requires selecting an appropriate size L . In any case, the Fourier transform of the resulting d -dimensional array is calculated numerically and then multiplied by $\sqrt{S_d(\omega)}$, and the inverse Fourier transform of the results is then computed numerically. The array so obtained follows the statistics of a FBM. To avoid the problem associated with the periodicity of the numbers arising as a result of their Fourier transforming, one must generate the array using a much larger lattice size than the actual size that is to be used in the analysis, and use the central part of the array. Clearly, a similar method can be used for generating a FGN. It should be emphasized that, as Hough (1989) pointed out, interpreting a power-law spectral density as an indication of fractality is not without difficulties and, thus, one must be careful in using such an analysis. In particular, a power-law spectral density might also be the characteristic of a nonstationary, but also nonfractal system.

The two-point correlation function $C_2(r)$ of a FBM array is given by

$$C_2(r) = C_0 r^{2H}, \quad (5.49)$$

which is similar to its semivariogram, Eq. (5.38), where, $C_0 = C(r = 1)$. To make the generation of a FBM array more efficient and accurate, Pang *et al.* (1995) and Makse *et al.* (1996c) modified the FFT method. Since the correlation function Eq. (5.49) has a singularity at $r = 0$, they considered a slightly different correlation function,

$$C_2(\mathbf{r}) = \left(1 + \sum_{i=1}^d r_i^2 \right)^{-\frac{\zeta}{2}}, \quad (5.50)$$

which, in the limit $r \rightarrow \infty$, has the same qualitative behavior as Eq. (5.49). The FT of the correlation function Eq. (5.50) can be determined analytically, with the result being,

$$S(\omega) = \frac{2\pi^{\frac{d}{2}}}{\Gamma(\beta + 1)} \left(\frac{\omega}{2} \right)^\beta K_\beta(\omega). \quad (5.51)$$

Here, $\beta = 1/2(\zeta - d)$, $\omega = |\omega|$, and K_β is the the modified Bessel function of order β . For $\omega \ll 1$, one has the asymptotic relation that $K_\beta(\omega) \sim \omega^\beta$. The

correlated array generated based on Eqs. (5.50) and (5.51) corresponds to a FBM with the Hurst exponent, $H = 1 - 1/2\xi$. Let us mention that Mehrabi and Sahimi (2009) extended the FFT method significantly in order to generate the FBM or FGN in a *curved* space, for example, along the curved strata in a FS porous medium.

5.5.2

Successive Random Additions

In the successive random addition method (Voss, 1985), one begins with the two end points in the interval $[0, 1]$, and assigns a zero value to them. Then, Gaussian random numbers Δ_0 with a zero mean and unit variance are added to these values. In the next stage, new points are added at a fraction r of the previous stage by interpolating between the old points (by either linear or spline interpolation), and Gaussian random numbers Δ_1 with a zero mean and variance r^{2H} are added to the new points. Thus, given a sample of N_i points at stage i with resolution λ , stage $i + 1$ with resolution $r\lambda$ is generated by first interpolating $N_{i+1} = (N_i - 1)(1/r - 1)$ new points from the old ones, and then adding Gaussian random numbers Δ_i to all of the new points. At stage i with $r < 1$, the Gaussian random numbers have a variance, $\sigma_i^2 \sim r^{2iH}$, consistent with the FBM. For example, with $r = 1/3$ and $N_i = 5$, the old (o) and new (n) points are in the order, (onnonnonnonno), so that there are $N_{i+1} = 8$ new points in the array. The process is continued until the desired length of the data array is reached. The method can easily be extended to any dimension (Lu *et al.*, 2003). It is not, however, very efficient if large 3D correlated arrays are to be generated. In addition, extending the method for generating *anisotropic* 2D or 3D arrays is difficult.

The problem with the method is that the points that are generated in earlier generations are not statistically equivalent to those generated later. To remedy this, one can add a random Gaussian displacement with a variance $r^{2(n-1)H}$ to *all* the points during the n th stage of the process, though doing so increases the computation time – it roughly doubles it. Moreover, if one is interested in generating an FBM array with a very wide range, one may start the process by assigning a Gaussian random number with a variance 2^{2H} to one end of the $[0, 1]$ interval. The generalization of this method to higher dimensions is straightforward.

5.5.3

The Wavelet Decomposition Method

The wavelet decomposition method is a suitable tool for analyzing the FBM-type data (Flandrin, 1992). For such data, one computes the following quantity,

$$D_j(k) = 2^{-\frac{j}{2}} \int_{-\infty}^{\infty} B_H(x) \psi(2^{-j}x - k) dx, \quad (5.52)$$

where $D_j(k)$ is called the *wavelet-detail coefficient* of the FBM, and represents the wavelet transformation of the data set. Here, ψ is the wavelet function (see Press *et*

al., 2007) for an introduction to wavelet functions) $k = 1, 2, \dots, N$, where N is the size of the data array, and the j s are integer numbers. Thus, in this method, one fixes j and varies k to calculate $D_j(k)$. For each j , one determines N such numbers and calculates their variance $\sigma^2(j)$. Then, it can be shown that regardless of the wavelet ψ , one has

$$\log_2[\sigma^2(j)] = (2H + 1)j + \text{constant} . \quad (5.53)$$

Thus, plotting $\log_2[\sigma^2(j)]$ versus j yields H .

The analysis of the porosity log of Figure 5.2 by the wavelet decomposition method is shown in Figure 5.3 (Sahimi and Mehrabi, 1999), which indicates that there are actually two distinct scaling regimes corresponding, respectively, to small (small j) and large (large j) length scales. For small length scales, one obtains $H \simeq 0.8$, indicating positive correlations. Such correlations are expected, as two points that are at short distances from each other presumably fall (at least in this case) within a single stratum, hence exhibiting positive correlations. On the other hand, for large length scales, one obtains $H \simeq 0.33$, indicating negative correlations, which is again expected as two points that are widely separated presumably belong to two strata with contrasting properties, hence exhibiting negative correlations.

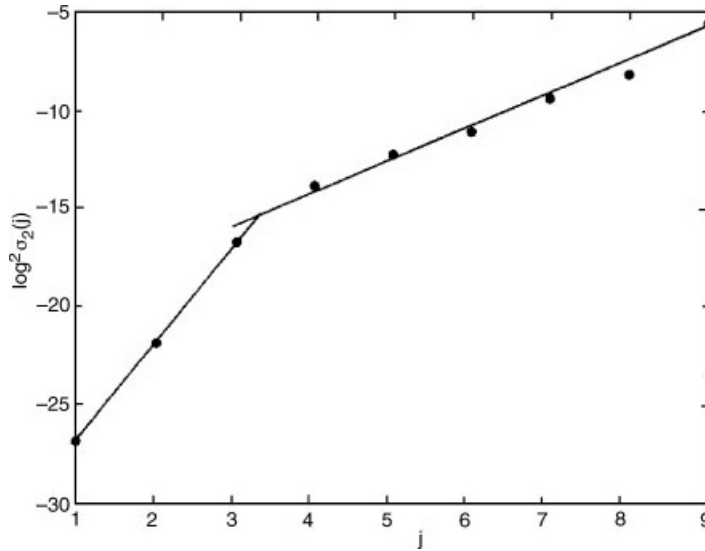


Figure 5.3 Wavelet decomposition analysis of the porosity log of Figure 5.2 (after Sahimi and Mehrabi, 1999).

5.5.4

The Maximum Entropy Method

The maximum entropy method computes the spectral density of the data *without* using the FFT, hence avoiding the pitfalls that one encounters in using the FFT. In this method, the spectral density is approximated by

$$S_d(\omega) \simeq \frac{a_0}{\left| 1 + \sum_{k=1}^M a_k z^k \right|^2}, \quad (5.54)$$

where the coefficients a_k are calculated such that Eq. (5.54) matches the Laurent series, $S_d(\omega) = \sum_{-M}^M b_i z^i$. Here, z is the frequency in the z -transform plane, $z \equiv \exp(2\pi i \omega \Delta)$, and Δ is the sampling interval in the real space. In practice, to calculate the coefficients a_i , one first computes the correlations functions

$$C_j = \langle d_i d_{i+j} \rangle \simeq \frac{1}{N-j} \sum_{i=1}^{N-j} v_i v_{i+j}, \quad (5.55)$$

where N is the number of data points, and d_i is the datum at point i . The coefficients a_i are then calculated from

$$\sum_{j=1}^M a_j C_{|j-k|} = C_k, \quad k = 1, 2, \dots, M. \quad (5.56)$$

The advantage of Eq. (5.54) over the Laurent series is that if $S_d(\omega)$ contains sharp peaks, it detects them easily because they manifest themselves as the poles of the equation, whereas one may have to use a very large number of terms in the Laurent series to detect the same peaks.

The analysis of the porosity log of Figure 5.2 by the maximum entropy method is shown in Figure 5.4 (Sahimi and Mehrabi, 1999). Except for large values of ω , that is, very short distances, the spectral density of the porosity log exhibits clear power-law scaling with the frequency. The estimated value of H , which is about 0.35, agrees with what one obtains with the wavelet decomposition method for large length scales. However, the spectral density method (and, hence, the maximum entropy method by which one computes the spectral density) is often not capable of detecting a scaling regime for short length scales, if one exists, because the measurement resolution is not high enough for distinguishing the points that are in the same neighborhood and, therefore, the data might be too noisy for the spectral density method.

While the four methods described are all highly accurate in terms of the minimum required size of the data array for reliable characterization of the long-range correlations, the maximum entropy method is the most reliable method (Mehrabi *et al.*, 1997) since it yields valuable information and accurate estimates of H , even when one has a small data array. This is a great advantage of the method since the development of a large data base for the FS porous media is costly and time consuming.

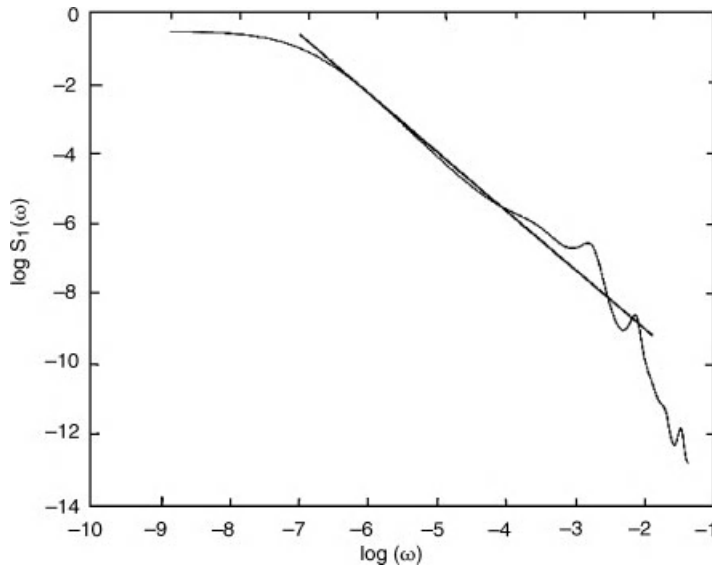


Figure 5.4 Spectral density $S_1(\omega)$ of the porosity log of Figure 5.2 computed by the maximum entropy method (after Sahimi and Mehrabi, 1999).

5.6

Interpolating the Data: Kriging

The analysis of the various well logs and other types of data is an important step toward characterization and modeling of the FS porous media. An even more important issue is how to interpolate and extrapolate the known data which are usually for the areas along the wells in, for example, oil reservoirs, to the interwell zones. Aside from some information for the large-scale structure of porous media that one gleans from seismic recordings, there are usually very little quantitative data for the interwell zones. In addition, even the data measured along the wells may contain significant gaps and, therefore, one must interpolate the existing data in order to fill the gaps.

This issue is particularly important because in order to develop the geological model of a FS porous medium, one must have quantitative information for every grid block of the model. However, the vast majority of such blocks represent the interwell zones and those representing along the wells where there are gaps in the existing data and, therefore, one must find an accurate way of interpolating and extrapolating the existing data to such zones. However, the problem sounds easier than it actually is. The problem is *uncertainty*, namely, given that a FS porous medium is typically highly heterogeneous, one must find a way of not only extrapolating the existing data to the zones for which there are no data, but also taking into account the effect of the uncertainty in any property value that is attributed to any grid block.

This becomes clearer once we recognize that every point in a FS porous medium as well as every random variable associated with the point (that represents a property value) has a PDF. If a property value at a given point is already measured – and, therefore, is known deterministically – then the PDF associated with it is simply a δ function or a spike because, aside from the possible measurement error, there is no uncertainty in the property value. If, however, a property value at that point has not been measured because, for example, it is impossible to do so, then a PDF is associated with it with a nonzero variance. If there are no physical constraints, for example, some qualitative information about the property, then the PDF can, in principle, take on any shape.

The best known technique for extrapolation and interpolation of the data is the *Kriging method*,²⁾ which is essentially a statistical method for estimating the property values for certain points, given some data for other points in the space. Krige (1951) developed a geological point of view that is based on the assumption of continued mineralization between measured values. Assuming, that is, prior knowledge encapsulates how minerals co-occur as a function of space. Then, given an ordered set of measured grades, interpolation by Kriging predicts mineral concentrations at unobserved points. Kriging has been used in mining, hydrogeology, modeling of fluid flow in oil reservoirs, natural resources, environmental science, remote sensing, and even the so-called black box modeling in computer experiments.

The theory of Kriging was developed by Matheron (1963, 1967a)³⁾, based on the Krige's work. Matheron had already derived an estimate K^* for the permeability which was the “estimateur” in his work and a precursor to the kriged estimate or kriged estimator. However, what he had failed to derive was $\sigma^2(K^*)$, the variance of his estimateur. In fact, he had computed the length-weighted average grade of each “panneau”, but did not compute the variance of its central value.

A central doctrine of the geostatistics that Matheron developed is that the spatial dependence (of some properties) need not be *verified*, but may be *assumed* to exist between two or more points, determined in samples selected at positions with different coordinates. This doctrine of assumed causality was the quintessence of Matheron's new geostatistics. The question remains, however, as to whether assumed causality makes sense in any other scientific discipline. For an account of the history of Kriging, see Cressie (1990).

The general idea that forms the basis for Kriging is as follows. Suppose that some data for a property, $Z_i = Z(x_i)$ with $i = 1, 2, \dots, N$, are known and one is interested in obtaining an estimate $\hat{Z}$ of the same property at a point for which

2) The method was first proposed by mining engineer *Daniel Gerhardus Krige* who was the pioneer plotter of distance-weighted average gold grades at the Witwatersrand reefs complex in South Africa. He developed the method as part of his Master's thesis, but never used the word *Kriging*.

3) *Georges Francois Paul Marie Matheron* (1930–2000) was a French mathematician and geologist who made important contributions to geostatistics and flow through porous media. The method was called *krigeage* for the first time in his 1960 *Krigeage un Panneau Rectangulaire par sa Périphérie*.

there are no data. A common interpolator is given by

$$\hat{Z} = \sum_{i=1}^N w_i Z_i . \quad (5.57)$$

Since the data points are spatial positions $x_1, x_2, \dots$, one may think of the parameters or parameters w_i as depending on the spatial position x_i as well. Formulating the estimator $\hat{Z}$ as in Eq. (5.57), the main problem is then determining the “optimal” values of the weights w_i , such that Eq. (5.57) provides an accurate estimate $\hat{Z}$. Note that Eq. (5.57) provides the estimate as a linear combination of all the known data Z_i . If the estimate is obtained by an unbiased method, namely, by one such that the expectation value of the property value is equal to its true value, then $\hat{Z}$ is called a BLUE (best linear unbiased estimate) estimate. We note that one may also use a *log-normal Kriging* that interpolates positive data by means of their logarithms. However, in what follows, we only work with the data themselves, rather than their logarithms.

Krige developed a method for estimating w_i , which is why the parameters are referred to as the Kriging weights (KWs). Depending on the stochastic properties of the random fields that represent the properties of FS porous medium, several types of Kriging have been developed. The type of Kriging used determines the linear constraint on the weights w_i implied by Eq. (5.57) and, hence, the method for calculating the weights depends upon the type of Kriging used.

5.6.1

Biased Kriging

This method is also called *simple Kriging*. Mathematically, it is the simplest of all the Kriging methods, but it is also the least general. It assumes that the expectation of the random field is known, and relies on a covariance function in order to determine the KWs w_i . However, in most applications, neither the expectation nor the covariance are known a priori. The method is built upon three fundamental assumptions: (1) the *wide sense* stationarity of the random field $Z(x)$; (2) the expectation of the field is zero everywhere, and (3) a covariance function is known. Thus, the method first defines a sensible variance. Let us define

$$\sigma_{BK}^2 = E[(Z - \hat{Z})^2] . \quad (5.58)$$

Note that the *Kriging variance* defined by Eq. (5.58) (as well as in what follows) must not be confused with the variance of the Kriging predictor itself. Equation (5.58), after substituting Eq. (5.57) for $\hat{Z}$, becomes

$$\sigma_{BK}^2 = \sigma_Z^2 - 2 \sum_{i=1}^N w_i R(Z, Z_i) + \sum_{i=1}^N \sum_{j=1}^N w_i w_j R(Z_i, Z_j) , \quad (5.59)$$

where $R(x, y)$ is the usual covariance of x and y . Note that the true value of the variance σ_Z^2 is not known. It is estimated from the sample variance or from the

semivariogram of the data (see Eq. (5.40)). To determine the KWs w_i , the variance is minimized with respect to w_i , that is, one sets $\partial \sigma_{\text{BK}}^2 / \partial w_i = 0$ for $i = 1, 2, \dots, N$. After taking the derivatives and some rearrangements, one obtains

$$\sum_{i=1}^N R(Z_i, Z_j) w_i = R(Z, Z_j), \quad j = 1, 2, \dots, N \quad (5.60)$$

which represents a set of N simultaneous linear equations for the N unknowns $w_1, \dots, w_N$. As the equations are linear, the minimum value of σ_{BK}^2 is actually its true global minimum, and not a local one.

Suppose that after determining the KWs w_i , we wish to obtain the Kriging estimates $\hat{\mathbf{Z}} = [\hat{Z}_1, \dots, \hat{Z}_n]^T$ at n grid points (T denotes the transpose operation). The estimates are given by the following vector equation:

$$\hat{\mathbf{Z}} = \mathbf{M}^T \mathbf{Z}, \quad (5.61)$$

where $\mathbf{M}$ is an $N \times n$ matrix of the KWs w_{ij} given by

$$\mathbf{M}^T = \begin{bmatrix} w_{11} & \cdots & w_{1n} \\ \vdots & \vdots & \vdots \\ w_{N1} & \cdots & w_{Nn} \end{bmatrix}. \quad (5.62)$$

Note that w_{ij} now has two indices which refer to the measurement (data) at point i and estimates at point j .

The resulting estimates are *biased* because in determining the KWs w_i , we did not impose the constraint that the true average value at the unsampled locations be produced.

5.6.2

Unbiased Kriging

Unbiased Kriging is also called *ordinary Kriging*, and is the most commonly used type of Kriging. It assumes a constant but unknown mean. Typical assumptions for using the ordinary or unbiased Kriging are (1) intrinsic or wide sense stationarity of the field $Z(x)$; (2) there are enough observations or data to estimate the variogram, and (3) the mean of the random field is unknown but constant. Under such assumptions, the KWs fulfill the unbiasedness condition. To remove the bias from the simple biased Kriging, we first recall Eq. (5.1) for the bias $b(\hat{\mathbf{Z}})$:

$$b(\hat{\mathbf{Z}}) = E(\hat{\mathbf{Z}}) - E(\mathbf{Z}) = E(\mathbf{Z}) \left(\sum_{i=1}^N w_i - 1 \right). \quad (5.63)$$

Thus, if $E(\mathbf{Z})$ is itself unbiased, a vanishing bias b requires setting

$$\sum_{i=1}^N w_i = 1. \quad (5.64)$$

In other words, the KWs w_i must be determined subject to the constraint that their sum must add to one. As such, determining the KWs for an unbiased estimate is a constraint optimization problem, a bit more difficult than determining the KWs w_i in the biased Kriging problem. To impose the constraints, one introduces the Lagrange multiplier χ in the variance to be minimized:

$$\sigma_{UK}^2 = \sigma_{BK}^2 + 2\chi \left(\sum_{i=1}^N w_i - 1 \right), \quad (5.65)$$

so that in addition to the KWs w_i , the Lagrange multiplier χ must also be determined. Use of the Lagrange multiplier ensures that the true minimum of σ_{UK}^2 is obtained. After minimizing σ_{UK}^2 with respect to both w_i and χ , one obtains

$$\sum_{i=1}^N R(Z_i, Z_j) w_i = R(Z, Z_j) + \chi, \quad j = 1, \dots, N \quad (5.66)$$

which is only slightly more complex than Eq. (5.59) and gives rise to a matrix equation similar to Eq. (5.60), but with one additional unknown, χ .

The main properties of any unbiased Kriging are as follows (Chilés and Delfiner, 1999; Wackernagel, 1995). (1) The Kriging estimates are unbiased. (2) The Kriging estimates honor the data, assuming that no measurement error exists. (3) The Kriging estimates are the best (most accurate) linear unbiased estimate if all the assumptions hold. If, however (Cressie, 1991), the assumptions do not hold, Kriging could result in poor estimates. (4) There might be better nonlinear and/or biased methods than Kriging. (5) No accurate estimate is guaranteed if the wrong variogram is used. However, one may still obtain a “good” interpolation. (6) The best estimate is not necessarily an accurate one. For example, if there are no spatial correlations, the Kriging interpolation is only as good as the arithmetic mean. (7) Kriging provides a measure of precision. The measure relies, however, on the correctness of the variogram.

5.6.3

Kriging with Constraints for Nonadditive Properties

The Kriging procedure described so far is usually accurate for obtaining estimates for those properties of a FS porous medium that are additive, such as the porosity. For nonadditive properties such as the permeability, however, even the unbiased constraint is weak. That is because an estimate of the permeability at any point in space is not normally a linear combination of the known (measured) permeabilities. Thus, one must impose more stringent constraints that result in relatively accurate estimates.

It is known that the average or overall permeability of a region that consists of several subregions with permeabilities $K_1, K_2, \dots$ satisfy certain upper and lower

bounds. For example, K satisfies the following bounds

$$\sum_{i=1}^N K_i^{-1} \leq K \leq \sum_{i=1}^N K_i . \quad (5.67)$$

Much tighter bounds are described by Sahimi (2003a). Thus, one may impose the constraint that the bounds Eq. (5.67) must be satisfied. Alternatively, one can use the results of well testing to constrain the permeabilities. In well testing, one measures the time-dependence of the variations in the pressure at the well by imposing changes on the volume flow rate of the well, from which the permeability of the porous formation around the well is estimated. Thus, one can constrain the estimate of the permeability based on well-testing results, although such a constraint is unable to provide information on the spatial variations of the permeability in the interwell zone. We should keep in mind that any nonlinear constraint comes at the price of more intensive computations.

5.6.4

Universal Kriging

Universal Kriging assumes a general linear-trend model. When there are long-range correlations in the data, then, in order to produce reasonable estimates by Kriging, one first subjects the data to a preprocessing step. For example, given some insight into the structure of a porous formation, one may divide the data into several segments, with each segment corresponding to a formation's zone, and then carry out separate Kriging for each zone.

5.6.5

Co-Kriging

If more than one set of data are available, then, in order to obtain Kriging estimates, one carries out a co-Kriging procedure. For example, suppose that in addition to a data set for the permeability along a certain path, additional information on the resistivity is also available, and we wish to obtain estimates of the permeability at the points for which no data are available. The estimate is then written as

$$\hat{Z} = \sum_{i=1}^N w_i Z_i + \sum_{i=1}^N \lambda_i X_i , \quad (5.68)$$

where $X_i = X(x_i)$ represent the additional data at x_i . Clearly, one may use exactly the same procedure that was described for the unbiased Kriging, except that one must determine two sets of co-Kriging weights, w_i and λ_i .

5.7

Conditional Simulation

Despite its many desirable features, Kriging is also not without problems. For one thing, the FS porous media are highly heterogeneous and, therefore, the spatial variations of such properties as the permeability are great – often by several orders of magnitude. However, the estimates that Kriging provides give rise to permeability fields that are too smooth. Sharp changes are almost never produced by Kriging alone. The second shortcoming of Kriging is that it is a completely deterministic method. In other words, the same data $Z(x_i)$ always produce the same KWs w_i and, hence, the same estimates $\hat{Z}$. The third significant feature of Kriging that can be bothersome is that all the estimates that it provides are Gaussian. This is easily seen by considering Eq. (5.57) and recalling the central-limit theorem.

The Gaussianity of the Kriging estimates is dealt with through a procedure called *disjunctive Kriging*. In this method, the data are preprocessed to be rendered Gaussian by a suitable transformation. The questions of smoothness of the fields generated by Kriging and their deterministic nature are addressed by *conditional simulation*. There are actually three variations on the conditional simulation, and what follows is a description of each.

5.7.1

Sequential Gaussian Simulation

As its name suggests, sequential Gaussian simulation (SGS) is a Kriging-based method that generates a Gaussian field for the entire system. The entire procedure can be summarized as follows (Jensen *et al.*, 2000). Suppose that we have N data points, $Z(x_i)$ with $i = 1, 2, \dots, N$, and wish to generate property values for M grid blocks, numbered $j = 1, 2, \dots, M$, including those at which the data are available. Thus, there are $M - N$ grid blocks that are *unconditioned*, that is, no data for them are available. We also assume that the semivariogram model is available as it can be computed using the N data points. The data are also assumed to be error free. The property values of the *conditioned* blocks, that is, those for which the data are available, do not change during the SGS.

1. The data are first transformed to be Gaussian.
2. The $M - N$ unconditioned grid blocks are assigned property values equal to those of the nearest conditioned blocks.
3. A random walk through the unconditioned blocks is constructed such that each of such blocks is visited only once.
4. Each time an unconditioned block is visited, the number and locations of the *local* conditioned blocks around it are identified. To do so, one must specify the shape and boundary of the region that constitutes the local neighborhood of the visited unconditioned block. Typically, the local neighborhood is defined in such a way that it has roughly the ellipse range identified by the semivariogram model (recall that most FS porous media are anisotropic). The local

- neighborhood may contain the blocks that have already been simulated, that is, a property value has already been assigned to them.
5. Unbiased Kriging (or co-Kriging if needed) is carried out for the local neighborhood using the data (actual as well as the already simulated that may be in the local neighborhood) as the conditioning points. Once this is done, the mean and variance σ_{UK}^2 of the local region are computed. The estimate $\hat{Z}$ has a Gaussian PDF with the computed mean and variance. Local Kriging is used in order to avoid solving the large-scale problem indicated by Eq. (5.65). However, one may also use a single Kriging for all the unconditioned grid blocks at once, thereby including all the trends at all the data points.
 6. A random number r , uniformly distributed in $[0, 1]$, is generated and used to pick a property value p from the Gaussian PDF constructed in step (v) by solving the equation

$$r = \int_{p_{\min}}^p f(x) dx ,$$

- where $f(x)$ is the Gaussian PDF, and $p_{\min}$ is the minimum value of the property. The value is attributed to the unconditioned grid block. Thus, the number of the data points (measured as well as simulated) increases by one.
7. The next step of the random walk to the next unconditioned grid block is taken and steps 4–6 are repeated.
 8. Steps 6 and 7 are repeated until all the unconditioned grid blocks are visited and property values are attributed to them. The final result is a *realization* of the model conditioned upon the available data.

Clearly, multiple realizations of the model can also be generated using this procedure. Each realization is a valid representation of the porous formation to be modeled.

5.7.2

Random Residual Additions

The random residual additions (RRA) method is, in some sense, more realistic than the SGS method because it no longer uses the concept of local neighborhood around an unconditioned grid block. What follows is a step-by-step description of the method.

1. An unbiased Kriging is carried out for the entire grid using the available data as the conditioning points.
2. An unconditioned grid is also generated using the semivariogram. Because it is unconditioned, it does not honor the data at the specified point. However, it does have all the required statistical properties, including the second moment of the PDF of the data because the semivariogram is used. The unconditioned system is *not* deterministic, but represents a single realization of the ensemble

of all the possible realizations with the required statistical properties. The unconditioned grid is generated by any appropriate method. For example, if the data follow a fractional Brownian motion (FBM), any of the methods of generating a FBM described earlier may be used.

3. Use the result of step 2 at the blocks for which the actual data are available as the conditioning points in order to carry out a second unbiased Kriging (or co-Kriging).
4. The unconditioned grid values of step 2 are subtracted from those generated by the second conditioned Kriging (or co-Kriging). Clearly, since the second Kriging was carried conditioned upon the unconditioned grid, the residuals for the blocks at which actual data are available are zero.
5. The resulting residuals obtained in step 4 are then added to the first Kriging carried out in step 1. Thus, the data are honored at the specified points – zero residuals are attributed to such grid blocks – and the nonzero residuals represent the uncertainty for the property values at the unconditioned grid blocks.

The resulting distribution of the property values in the computational grid has many desirable features: (1) it honors the data at the specified points; (2) it has the correct statistical properties; (3) it is unbiased, and (4) it includes the large-scale heterogeneity that is expected to exist in the geological formation and its model.

5.7.3

Sequential Indicator Simulation

This is a method that classifies each grid block into a facies category. The category is built up using a variety of data, such as, seismic-amplitude map extractions that are calibrated using core and log data and include, for example, channels, overbank deposits, and others.

It is assumed that no two facies can exist in the same grid block. Thus, the method is sometimes referred to as a simulation for generating the facies. After each grid block is assigned to its facie category, its property value is attributed to it from the PDF of the facie. Thus, the method is also called *sequential indicator simulation-probability distribution function* (SIS-PDF). The overall PDF of the facies represents the pattern of their occurrence at the scale of the porous medium to be modeled. The PDF is obtained from the usual sources, that is, either log data or a map of the facies. The overall procedure for the SIS-PDF is as follows (Jordan and Goggin, 1995).

1. As in the SGS method, a random walk is taken through the computational grid that represents the FS porous medium such that the unconditioned blocks are visited once and only once.
2. For each visited unconditioned grid block, the prespecified number of conditioning facies data from the wells and already simulated blocks, as well as any other sources of data, is identified.

3. An *indicator Kriging* is carried out in order to estimate the conditional probability for each facies category. An indicator Kriging uses indicator functions instead of the process itself in order to estimate the transition probabilities. It proceeds exactly like the usual Kriging, except that an *indicator* semivariogram γ_I is used in place of the usual semivariogram, and the data values Z_i are replaced by their respective indicators. To construct γ_I , a threshold value Z_c is introduced that varies between a minimum and maximum value and, hence, can take on several values Z_{c_i} . Then, the indicator variable $I(Z_{c_i})$ is defined by

$$I(Z_{c_i}) = \begin{cases} 1 & Z_i \leq Z_{c_i} , \\ 0 & Z_i > Z_{c_i} . \end{cases}$$

The cumulative distribution function $\hat{F}(Z_c)$ is then constructed by

$$\hat{F}(Z_c) = \frac{1}{M} \sum_{i=1}^M I(Z_{c_i}) ,$$

where $M < N$, with N being the number of the data points. The indicator semivariogram is constructed based on $I(Z_{c_i})$, and the cumulative probability distribution is used for estimating the conditional probabilities.

4. Each facie's probability is then normalized by the sum of the probabilities of all facies. The result is then used to construct a local cumulative probability distribution.
5. A random number r , distributed uniformly in $[0, 1]$, is generated and used together with the local cumulative distribution function in order to determine the simulated facies category in the visited unconditioned grid block.
6. For each unconditioned block in the random path, steps 2–5 are repeated. The final result is the facies distribution.

5.7.4

Optimization-Reconstruction Methods

In addition to Kriging and conditional simulation methods, there are other techniques that have been used for developing geological models of the FS porous media. They do not, however, represent geostatistical methods and have been borrowed from other disciplines, for example, the statistical mechanics of thermal systems. Chief among them, are the simulated annealing method and the genetic algorithm, both of which represent optimization methods. That is, given a limited amount of data, the two methods are used for determining an optimal structure (spatial distributions of the porosity and permeability) of a porous medium that not only honors the data, but can also provide accurate predictions for other properties for which no data are available. This method is also referred to as *reconstruction*, as one attempts to reconstruct a model of a porous medium for which limited amounts of data are available. The two optimization methods will be described in detail in Chapters 6–8.

6

Characterization of Fractures, Fracture Networks, and Fractured Porous Media

Introduction

Among the many problems that must be addressed by chemical and petroleum engineers who deal with oil and gas reservoirs, and by hydrologists and hydrogeologists who work on the problem of groundwater contamination, none is perhaps as difficult as the characterization and modeling of fractures, fracture networks, and fractured porous media in which oil, gas, and groundwater reside and flow. For example, the economic development of a fractured oil reservoir – which are abundant in the world – cannot be planned unless the nature of the fractures (mechanical, or diagenetic, or both) and the structure of their network (connectivity, fracture length distribution, etc.) are first understood, and accurate models for both the fractures and their network are developed. Similar problems arise in hydrology when one wishes to address the problems of groundwater flow and its contamination.

Most recently, the problem of disposal of radioactive waste has become a critical problem of national and international concern. Geological formations have emerged as an acceptable place for storing the radioactive nuclear wastes since the permeability of such formations is typically low. However, many geological formations that can potentially be used for this purpose are highly fractured, even several hundreds of meters below the surface. Groundwater that flows through the fractures at such depths would be the most important carrier of any nuclear waste and contaminants that might leak out of the storage area and enter the flow system (Kersting *et al.*, 1999). Hence, characterization and modeling of fractured rock has, once again, emerged as a fundamental problem of immense importance.

Despite years of study, the problem of accurate identification of fractures' spatial distribution remains an active area of research. Although the permeability of fractures is much larger than that of the porous matrix in which they are embedded and, thus, permeability data measured at various points can provide insight into the spatial distribution of fractures, there is usually an insufficient volume of such data. Borehole-wall imaging is a very reliable method of mapping the fractures and faults within boreholes. However, such imaging techniques are expensive and are not always included in a logging run. In addition, such images are not available for, for example, many of the older oil reservoirs.

The purpose of this chapter is to describe characterization and modeling of fractures and fracture networks. Although the subject has been studied for several decades, over the past 25 years or so, many new techniques and models have been developed for characterization of fractures and fracture networks. At the same time, the improved understanding that has been gained is instrumental to obtaining improved predictions for flow and transport properties of field-scale (FS) porous media. Any discussion of characterization fractures and fractured porous media is usually divided into three distinct, but related, parts that are related to the characterization of

1. a single fracture;
2. a network of fractures, and
3. a fractured porous medium.

In the third case, the fractures may be *discretely* present. That is, they do *not* form an interconnected network between two widely-separated parts of a porous medium or, for example, between the injection and extraction wells in an oil reservoir, although they may form local clusters of interconnected fractures. Alternatively, the fractures may form a network at large length scales, making it the main conduit for flow and transport processes.

It has been established over the past 20 years that many properties of fractures and fracture networks follow power-law distributions, characterized by long-range correlations. In addition, it now appears that the connectivity of fracture networks and its effect on the network's properties may be accurately quantified by the concepts of percolation theory described in Chapter 3, while the internal surface of fractures, which is typically very rough, may be understood and modeled in terms of self-affine stochastic functions. The goal of this chapter is to describe such developments.

6.1

Surveys and Data Acquisition

Before starting our main discussions, let us briefly describe how experimental data for various characteristics of fractures and fracture networks are obtained. There are three types of *surveys* for characterizing rock:

1. In a *borehole survey*, the measurements are usually made on oriented cores. Alternatively, they can be deduced from the resistivity of various samples (resistivity logs). The location, orientation, aperture, surface geometry, and other properties are recorded for each fracture. This type of survey provides information about the fracture density and its regionalization, the orientation distribution, and the presence of fracture sets. For application to oil and geothermal reservoirs see, for example, Narr and Lerche (1984) and Genter *et al.* (1995).

- Borehole surveys do not, however, provide any insight into the extent of the fractures or their interconnectivity, as they are local.
2. A *scanline survey* is an extension of borehole surveys in which one paints and collects the fractures that intersect a horizontal line on an outcrop. Each fracture is represented by its trace. The intersection abscissae, and trace lengths and orientations may also be recorded (Priest and Hudson, 1981). This survey is usually used for pit batters and high bench walls where it is possible to observe long traces.
Sisavath *et al.* (2004) developed a method by which several important geometrical and connectivity properties of fracture networks can be derived from line data. They showed that when data relative to fractures are collected along a line, such as a road or well, estimates of major geometrical properties of the corresponding fracture networks, for example, the volumetric density of fractures and their percolation properties, can be obtained.
 3. In an *areal survey*, one collects all the fracture traces that intersect a rectangle. If the traces continue beyond the rectangle, only that portion of them that is within the rectangle is recorded. This type of survey is usually used for drift or tunnel walls (see, for example, Rouleau and Gale, 1987; Billaux *et al.*, 1989; Abelin *et al.*, 1991), but can also be used for outcrops. Odling (1992) studied an 18×18 m area from ground observations; Koestler and Reksten (1992) mapped a 45×230 m quarry well, and Barthélémy *et al.* (1996) used photographs taken from aircrafts or helicopters. Vignes-Adler *et al.* (1991) even used images obtained by satellites. Castaing *et al.* (1996) utilized a variety of techniques in order to characterize a fractured area in the Western Arabian Plate. The sampling scales used were anywhere from 10 m to as large as 1000 km.

Thus, although sampling of the spatial properties of fracture networks should, in principle, be three-dimensional (3D), the surveys described here are essentially 1D or 2D since 3D surveys and samplings are difficult. They require geophysical imaging and mapping of the fracture surfaces in a volume of the rock. One can interpolate between a sequence of closely spaced 2D fracture-trace maps that are parallel to one another.

Full 3D characterization of fracture networks is very difficult and, therefore, very few attempts have been made in this direction. Montemagno and Pyrak-Nolte (1995) and Pyrak-Nolte *et al.* (1997) used X-ray computed tomography in order to characterize small samples. Gertsch (1995) and Gonzalez-Garcia *et al.* (2000) constructed 3D fracture networks from serial sectioning data (see Chapter 4). We should also mention the work of Crosta (1997) who provided practical guidelines for ground-level data acquisition.

6.2

Characterization of Surface Morphology of Fractures

Similar to unfractured porous media, natural fractures and their networks have enormous variations in their morphology that consists of the geometry, topology and the structure of the internal surface of the fractures. The geometry refers to size or aperture of fractures as well as their shape. The topology describes how fractures are connected to one another. The structure of the internal surface of fractures is also very important because it is often very rough and possesses complex features. Indeed, as we describe below, it has been shown that the internal surface of fractures (as well as that of pores; see Chapter 4) is very rough, but that the roughness is not random. Rather, as described shortly, it is often described by self-affine stochastic functions, hence implying the existence of extended correlations in the surface structure. The roughness of a fracture's internal surface affects, of course, its flow and transport properties, and will be studied in Chapter 12.

However, when we speak of surface roughness, we must specify the length scales over which the roughness is measured. Even the most rugged mountains look perfectly smooth when viewed from the outer space! Therefore, surface roughness and, more generally, all the morphological characteristics of fractures, fracture networks, and fractured porous media *depend on the length scale of observations or measurements*. The effect of the topology of disordered fracture networks on their effective flow and transport properties is quantified by percolation theory.

Although the enormous variations in the morphology of fractures and fractured porous media, particularly in their surface structure, are such that, up until the mid 1980s, the problem of describing and quantifying such morphologies seemed hopeless, significant experimental and theoretical developments have brightened the prospects for deeper understanding of the morphology, and in particular the structure of their internal surface. Among them are the advent of powerful concepts of fractal geometry and fractal distributions. Such ideas have advanced our understanding of the connectivity and surface structure of fractures. As described in Chapter 5, fractal distributions play a fundamental role in describing many properties of FS porous media. As described below, fractal concepts provide us with a powerful tool for characterizing the structure of a fracture's internal surface and its roughness.

6.2.1

Self-Similar Structures

An intuitive and informal definition of a self-similar fractal object was already given in Section 3.8: In a self-similar object, the part is reminiscent of the whole, implying that the object possesses *scale-invariant* properties. That is, the object's morphology repeats itself at different length scales. This means that above a certain length scale – the lower cutoff scale for fractality – the structure of a piece of an object can be magnified to recover its structure at larger length scales up to another length scale – the upper cutoff for its fractality. Below and above the two

cutoff length scales, the system loses its self-similarity. While there are disordered media that are self-similar at *any* length scale, natural porous media that exhibit self-similarity typically lose their fractal characteristics at sufficiently small or large length scales.

As already described in Chapters 3 and 4, one of the simplest characteristics of a self-similar fractal system is its *fractal dimension* D_f , which is defined as follows. We cover the fractal system by non-overlapping d -dimensional spheres of Euclidean radius r , or boxes of linear size r , and count the number $N(r)$ of such spheres that completely cover the system. The fractal dimension D_f of the system is then defined by

$$D_f = \lim_{r \rightarrow 0} \frac{\ln N}{\ln \left(\frac{1}{r} \right)}. \quad (6.1)$$

Estimating the fractal dimension through Eq. (6.1) is called the *box-counting method*. For non-fractal objects, $D_f = d$, where d is the Euclidean dimensionality of the space in which the objects are embedded. Note that to invoke Eq. (6.1), we implicitly assume the existence of a lower and an upper cutoff length scale for the fractality of the system that are, respectively, the radius r of the spheres and the linear size L of the system.

One may also define the fractal dimension D_f through the relation between the system's mass M and its characteristic length scale L . If the system is composed of particles of radius r and mass m , then

$$M = cm \left(\frac{L}{r} \right)^{D_f}, \quad (6.2)$$

where c is a geometrical constant of order 1. Since we can fix the dependence of M on m and r , we can write

$$M(L) \propto L^{D_f}. \quad (6.3)$$

Measuring M often entails using an ensemble of samples with similar structures, rather than a single sample. In this case,

$$\langle M \rangle = cm \left(\frac{L}{r} \right)^{D_f}, \quad (6.4)$$

where $\langle \cdot \rangle$ indicates an average over the mass of a large number of samples with linear sizes in the range $L \pm \delta L$, centered on L . Natural fractals are usually *statistically self-similar* because their self-similarity is only in an average sense. For example, if a fracture network or a cluster of fractures has a self-similar structure, its self-similarity is only in an average sense. No exactly self-similar fracture network is known to exist in nature.

6.2.2

The Correlation Functions

A powerful method for testing self-similarity of any disordered medium is to construct a general correlation function $C_n(\mathbf{r}^n)$ defined by

$$C_n(\mathbf{r}^n) = \langle \rho(\mathbf{r}_0) \rho(\mathbf{r}_0 + \mathbf{r}_1) + \dots + \rho(\mathbf{r}_0 + \mathbf{r}_n) \rangle, \quad (6.5)$$

where $\rho(\mathbf{r})$ is the density at position $\mathbf{r}$, and the average is taken over all possible values of $\mathbf{r}_0$. Here, $\mathbf{r}^n$ denotes the set of points at $\mathbf{r}_1, \dots, \mathbf{r}_n$. C_n , in the limit $n = 2$, is usually referred to as the covariance function. If an object is self-similar, then its correlation function defined by Eq. (6.5) should remain the same (aside from a constant prefactor) if all the length scales of the system are rescaled by a constant factor b . Thus, one must have

$$C_n(b\mathbf{r}_1, b\mathbf{r}_2, \dots, b\mathbf{r}_n) = b^{-nx} C_n(\mathbf{r}_1, \dots, \mathbf{r}_n). \quad (6.6)$$

Only a *power-law* correlation function can satisfy Eq. (6.6) with $x = d - D_f$, where x is called the *co-dimensionality*. In most cases, however, only the two-point correlation function $C_2(\mathbf{r}^2)$ – also called the *direct correlation function* or the covariance function – can be computed or measured with high precision and, therefore, we focus on this quantity. Assuming isotropy, $C_2(\mathbf{r}^2)$ is the same as the correlation function $C(r)$, introduced by Eq. (4.105). Then, due to self-similarity of the system, the direct correlation function $C(r)$ *decays* as a power law, given by Eq. (4.106). Thus, a logarithmic plot of $C(r)$ versus r should be a straight line with a slope $D_f - d$. Equation (4.106) has an important implication: There are long-range correlations in the morphology of a self-similar fractal system that decay slowly as $C(r) \rightarrow 0$ only when $r \rightarrow \infty$.

6.2.3

Rough Self-Affine Surfaces

The self-similarity of a fractal structure implies that its microstructure is invariant under an *isotropic* rescaling of lengths, that is, if all lengths in all directions are rescaled by the same scale factor, the same structure is obtained again. There are, however, many fractals that preserve their scale-invariance only if lengths in different directions are rescaled by direction-dependent scale factors. In other words, their scale-invariance is preserved only if the lengths in the x , y , and z directions are scaled by scale factors b_x , b_y , and b_z , where, in general, the scale factors are not equal. This type of scale-invariance implies that the fractal system is, in some sense, *anisotropic*. Such fractal systems are called *self-affine*. A self-affine fractal object can no longer be described by a single fractal dimension D_f . A simple example of a self-affine fractal is the curve in the (x, t) plane, generated by the equation, $x = F(t)$, where $F(t)$ is the displacement of a particle undergoing 1D diffusion or Brownian motion. In this example, for any $b > 0$, $F(t)$ and $b^{-1/2}F(bt)$ are statistically identical. To see this, recall that the probability density function $P(x, t)$ for

finding a diffusing particle at position x at time t satisfies the standard diffusion equation,

$$D \frac{\partial^2 P}{\partial x^2} = \frac{\partial P}{\partial t}, \quad (6.7)$$

where D is the diffusivity. It is not difficult to show that Eq. (6.7) remains invariant under the rescalings $(x, t) \rightarrow (b^{1/2}x, bt)$. Thus, the invariance is preserved by rescaling the x and t axes by different scale factors.

A well-understood process that gives rise to a self-affine fractal is a marginally stable growth of an interface. For example, if water displaces oil in a water-wet porous medium, the interface between water and oil is a self-affine fractal (see Chapter 13). Among natural self-affine surfaces are pores and fractures of rock. Many properties of such surfaces are described by a function $f(\mathbf{x})$ that also possesses a self-affine structure. For example, the surface height $h(x, y)$ at a lateral position $\mathbf{x}$ of the rough internal surface of a fracture has self-affine property (see below). In fact, self-affinity of many natural systems that are associated with Earth, such as various properties of rock, is quite understandable since gravity plays a dominant role in one direction but has very little, if any, effect in the other directions, hence generating anisotropy in the rock's structure.

Similar to self-similar structures, the self-affinity of the internal surface of fractures is only in a statistical sense. In addition, a disordered self-affine fractal surface may be thought of as the fluctuations about a straight line or a flat surface. Such fluctuations generate rough, self-affine profiles and surfaces. If we consider the height difference between a pair of points $h(\mathbf{x}_1)$ and $h(\mathbf{x}_2)$ on a self-affine surface $h(\mathbf{x})$ that lie above or below points separated by a distance $x_1 - x_2 = x = |\mathbf{x}|$ on a flat reference surface (or a line in 1D), then

$$\langle |h(\mathbf{x}_1) - h(\mathbf{x}_2)| \rangle \sim x^H, \quad (6.8)$$

where H is called the *roughness exponent*, often also called the *Hurst exponent*. Recall that the Hurst exponent was also utilized in Chapter 5 to characterize the porosity logs, permeability distributions, and many other properties of FS porous media. One may generalize Eq. (6.8) to higher dimensions and generate rough surfaces that are encountered in a variety of contexts, ranging from surface of pores of natural porous media (see Chapter 4) to the internal surface of fractures of rock (see below), fracture surface of heterogeneous materials, and thin solid films (Sahimi, 2003b).

6.2.4

Measurement of Surface Roughness

In practice, self-affinity of a surface is bounded by an upper correlation length ξ^+ and a lower correlation length ξ^- in both the horizontal ($\parallel$) and vertical ($\perp$) directions, hence restricted to the ranges, $\xi_{\parallel}^- < \delta x < \xi_{\parallel}^+$ and $\xi_{\perp}^- < \delta h < \xi_{\perp}^+$, where

h is the height of the surface. Then, due to self-affinity, we must have

$$\frac{\xi_{\perp}^{+}}{\xi_{\perp}^{-}} = \left(\frac{\xi_{\parallel}^{+}}{\xi_{\parallel}^{-}} \right)^H . \quad (6.9)$$

However, the numerical value of the Hurst or roughness exponent H is not sufficient for characterizing the roughness of a surface, as H only indicates how the roughness (or the variance in the height) varies with the transverse length scale over which it is measured. A complete characterization of a rough surface would require not only H , but also the *amplitudes* of the height fluctuations as well as the transverse correlation lengths. One way of characterizing a rough surface is by measuring the width w of the surface defined by

$$w(\ell) = \left\langle [h(\mathbf{x}) - \langle h \rangle_{\ell}]^2 \right\rangle^{\frac{1}{2}} , \quad (6.10)$$

where $h(\mathbf{x})$ is the height of the surface at position $\mathbf{x}$, and $\langle h \rangle_{\ell}$ is its average over a horizontal segment of length ℓ , normalized by the “volume” ℓ^{d-1} for a d -dimensional surface. Over a segment of size ℓ of the surface, and for $\ell \ll \xi_{\parallel}$, we must have

$$w(\ell) \sim \ell^H . \quad (6.11)$$

For $\ell \gg \xi_{\parallel}$, we must, of course, have $w = \xi_{\perp}$.

Another method of characterizing a rough surface is by the so-called *slit island* method (Mandelbrot *et al.*, 1984). In this method, the rough surface is coated with another material and then carefully polished parallel to the flat reference surface (described above) to reveal a series of horizontal cuts. As the coating material is removed, islands of the surface material appear in a sea of the coating material. With further removal of the coating material, the islands will grow and merge. If we consider a region of linear size ℓ and height fluctuations $\Delta h = h(\mathbf{x}) - \langle h \rangle_{\ell}$, the distribution $P(\Delta h)$ of the height fluctuations is described by the following scaling law

$$P(\Delta h) = w(\ell)^{-1} f \left[\frac{\Delta h}{w(\ell)} \right] . \quad (6.12)$$

Since $w(\ell)$ follows Eq. (6.11), the implication is that the density $\rho(\ell)$ in a cross-section of size ℓ must be given by

$$\rho(\ell) \sim \ell^{-H} . \quad (6.13)$$

Equation (6.13) suggests that the interface between the two materials, that is, between the rough surface and the coating material, in the cross-sections parallel to the reference plane is a *self-similar* fractal with a fractal dimension

$$D_f = d - H . \quad (6.14)$$

Therefore, if the fractal dimension D_f can be independently estimated, then the Hurst or roughness exponent H will also be evaluated. Typically, the islands that appear have a surface area distribution n_A such that

$$n_A \sim A^{-\tau}, \quad (6.15)$$

where n_A is the number of islands with areas A in the range $[A - \frac{1}{2}\Delta A, A + \frac{1}{2}\Delta A]$. The exponent τ is related to the fractal dimension D_f through the following equation

$$\tau = \frac{1}{d} (D_f + d), \quad (6.16)$$

so that measurement of the islands' areas yields D_f , from which the roughness exponent H is estimated.

A third method for characterization of a rough surface uses a mechanical stylus profilometer that provides, by design, 1D profiles of the surface heights. The resolution of the method is in the range 1–10 μm , and was probably used for the first time by Whitehouse and Archard (1970). Since their pioneering work, the method and instrument have been refined significantly (see, for example, Boffa *et al.*, 1998) by using a computer to control and precisely position the device on the surface. In addition, more advanced techniques based on scanning tunneling microscopy (Mitchell and Bonnell, 1990) and atomic force microscopy (Daguier *et al.*, 1996) have also been used for characterization of rough surfaces.

6.3

Generation of a Rough Surface: Fractional Brownian Motion

How does one generate synthetic (numerical) rough self-affine curves and surfaces? The generation is usually done by using a self-affine fractal function, the properties of which may also be used as a guide for better understanding of rough surfaces that one encounters in practical applications. In addition, because the stochastic self-affine process generates fractal sets with long-range correlations, it may also be used for modeling of a variety of phenomena in engineering and materials science in which the effect of such correlations is paramount. The stochastic fractal function that is usually used for generating rough self-affine surfaces is the fractional Brownian motion (FBM), already introduced in Section 5.5 for describing porosity logs, the permeability distributions, and other important properties of FS porous media. However, whereas the significance of the FBM described in Chapter 5 was that it described long-range correlations in the properties of FS porous media, in the present context it describes the morphology of rough self-affine surfaces. We already described the main properties of the FBM and FGN in Chapter 5. It should suffice to add that the covariance function $C(r)$ of a FBM is given by

$$C(r) - C(0) \sim r^{2H} \quad (6.17)$$

so that as long as values of $H > 0$ (the only physically-acceptable values for porous media), the correlations *increase* as r does. Efficient and accurate generation of a d -dimensional array that follows the statistics of a FBM (or FGN) is not straightforward, but was described in detail in Section 5.5.

6.4

The Correlation Function for a Rough Surface

How does one characterize a rough self-affine surface generated either synthetically or by a physical process? We define a height correlation function $C_h(\mathbf{x})$ by

$$C_h(\mathbf{x}) = \langle |h(\mathbf{x}_0 + \mathbf{x}) - h(\mathbf{x}_0)|^n \rangle^{\frac{1}{n}}, \quad (6.18)$$

where the averaging is over all the origins $\mathbf{x}_0$. The choice of the reference surface should be made carefully. For example, if the rough surface has been grown from a planar substrate, then the original flat surface would be the logical choice. In any case, it has been found for many rough surfaces that

$$C_h(\mathbf{x}) = \langle C_h(\mathbf{x}) \rangle_{|\mathbf{x}|=x} \sim x^{H(n)}, \quad (6.19)$$

where the averaging is taken with respect to all the origins $\mathbf{x}_0$ in the smooth reference plane. In most cases, the exponents $H(n)$ take on the same value H for all n . A surface with correlation function given by Eq. (6.19) is a self-affine fractal over the range of length scales in which $C_h(\mathbf{x})$ is computed. Typically, the case $n = 2$ has been utilized for estimating H , and has proven to be a very robust and accurate method. The correlation function $C_h(\mathbf{x})$ satisfies a general scaling equation given by

$$C_h(\mathbf{x}) = x^H F_n \left(\frac{x}{\xi_{\parallel}^+}, \frac{x}{\xi_{\parallel}^-} \right). \quad (6.20)$$

For $x \gg \xi_{\parallel}^-$, such that $x/\xi_{\parallel}^- \rightarrow \infty$, scaling Eq. (6.20) simplifies to

$$C_h(\mathbf{x}) = x^H f \left(\frac{x}{\xi_{\parallel}^+} \right), \quad (6.21)$$

where the scaling function $f(\gamma)$ has the properties that $f(\gamma) = c$ for $\gamma \ll 1$ and, $f(\gamma) \sim \gamma^{-H}$ for $\gamma \gg 1$, where c is a constant of order unity.

6.5

Characterization of a Single Fracture

A single fracture is characterized by several properties. Thus, in what follows, we consider each of such properties and describe their measurements.

6.5.1

Aperture

One of the most important properties of a single fracture is its aperture that controls its flow and transport properties. However, as described below, the internal surface of a fracture is typically rough. Therefore, there is a *distribution* of the apertures, rather than a unique aperture for a given fracture. The distribution can be obtained only if the fracture's internal surface is characterized, and the distribution of the contact areas in the interior of the fracture is obtained. In addition, a fracture is also characterized by its length and displacement – properties that manifest their effect in a network of interconnected fractures – and, therefore, the experimental data for such properties and their implications will be described below.

One way of determining the aperture is using the profilometer for the surface topographies to construct the void space of a single fracture (Keller and Bonner, 1985), although one must be careful when using the technique since many artifacts can affect the constructed structure of the void space. In addition, 2D cuts provide direct estimates of the aperture that is usually visible in such cuts, and are obtained either from natural outcrops or fracture traces, notwithstanding their possible changes by natural or human activities. To preserve the open space of a single fracture, one usually injects resin into the fracture. For example, Hakami and co-workers (Hakami and Stephansson, 1993; Hakami, 1995; Hakami and Larsson, 1996) injected polyurethane in fractures in granites. The aperture field is then reconstructed by image analysis of sections of the sample. Measurements were also carried out on transparent replicas of natural fracture (Yeo *et al.*, 1998; Koyama *et al.*, 2006; Bauget and Fourar, 2008). A dyed fluid was then injected into the void space of the fracture, a calibration law was established to relate the local light absorption to the local apertures, and the distribution of the local apertures was then measured.

Such techniques make it possible to construct the distribution of the apertures. Both normal (Vickers *et al.*, 1992; Hakami and Larsson, 1996; Yeo *et al.*, 1998; Matsuki *et al.*, 2006) and log-normal (Snow, 1970; Gale, 1982, 1987; Hakami, 1995; Pyrak-Nolte *et al.*, 1997; Keller, 1998; Koyama *et al.*, 2006) appear to represent the distribution accurately, hence indicating some dependence on the nature of the rock and its deformation mode. Log-normal distributions have been reported mostly for fractures that had been submitted to a normal stress. The reason is clear: with a normal stress, the apertures are shifted toward lower values because the surfaces come into close contact; see Oron and Berkowitz (1998) and Sharifzadeh *et al.* (2008) for more discussions.

The aperture changes under mechanical deformation. If a normal stress is applied to a fracture, contact areas emerge, the local apertures are reduced, and the deformation varies nonlinearly with the stress. The nonlinearity reflects the changes in the void space of the fracture. The rate of deformation is largest when small stresses are applied, which indicates that fracture stiffness has increased. Such phenomena were studied by Pyrak-Nolte *et al.* (1987) who used injection of an alloy with a low melting point into the fracture in order to obtain a cast of its void

space. Studies have also been reported in which the effect of shear displacements as well as that of various variations of normal stresses on the aperture distribution was studied. Such studies have demonstrated that deforming a fracture subject to a shear stress significantly changes its void space. In particular, two main features have been reported, namely, (1) *dilatancy* of the fracture (i.e., the rock *expands* as it is *squeezed*) that increases the apertures, and (2) the emergence of damage zones.

Therefore, what happens when a shear stress is applied to a fracture? At first, small variations of the apertures are observed up to a shear displacement of a few millimeters (see, for example, Sharifzadeh *et al.*, 2008). This is due to the fact that the motion is stopped when the asperities of the opposite surfaces of the fracture come into contact. Then, there is uplift of the two halves that reflect the dilation as the shear stress increases simultaneously up to a maximum value. Beyond this point, the shear stress rapidly decreases to a residual value that is essentially constant. At a critical value near the maximum shear stress, all the asperities in contact are simultaneously sheared, which results in a sudden change in the distribution of the apertures because the roughness of the two opposite surfaces no longer match. If the shear stress is increased further, the apertures increase that results in an increase in the fracture permeability by about one order of magnitude.

6.5.2

Contact Area

There has also been interest in determining the contact area in fractures under in-situ stress conditions. For example, Gentier *et al.* (1989) generated a cast of the open space of a granite fracture by injecting a dyed translucent resin into it which can be done if the sample is under normal stress as well as without any external load. They then removed half of the rock sample and counter-molded a second thicker layer of transparent resin onto the first one, which produced a composite cast with a thin layer of dyed resin that corresponded exactly to the void space of the fracture, and a transparent thicker layer acting only as the mechanical support of the system. The distribution of the apertures was then deduced from a gray-level image of the cast illuminated by transmission. The connectivity of all the void zones of the fracture can also be obtained from the skeleton of the open space.

Hakami and Barton (1990) and Hakami (1992) used a somewhat different approach by replicating both sides of several fractures and using them as molders to cast transparent replicas of the rock samples, which were then fitted together to produce a transparent sample of the original fracture. To measure the apertures, calibrated water droplets or soft rubber pieces were spread on the fracture's lower surface, and their apparent surface areas were measured when the fracture was closed.

All such methods suffer from one problem or another. Perhaps the most accurate and problem-free method for measuring the various characteristics of a single fracture is 3D X-ray computed tomography that allows measurements down to a few microns (Johns *et al.*, 1993; Spanne *et al.*, 1994; Keller, 1998; Arns *et al.*, 2001, 2002; Bertels *et al.*, 2001; Gouze *et al.*, 2003; see also Chapter 4). We will revisit the

question of the distribution of the contact area in Chapter 8, where we describe the percolation properties of fractures and fracture networks.

6.5.3

Surface Height

The key quantity to modeling a single fracture is its surface height or elevation $h(x, y)$ which controls the fracture aperture. The surface characteristics are expressed either in terms of various statistics of h itself, for example, its central-line average $\langle |h| \rangle$ (recall that, $\langle h \rangle = 0$) and variance $\sigma_h^2 = \langle h^2 \rangle$, or in terms of its derivatives, for example, $Z_2 = \langle \partial h / \partial x \rangle^2$. Moreover, in addition to the correlation function defined by Eqs. (6.18)–(6.21), the covariance function of h is also defined,

$$C_2(\mathbf{r}) = \langle h(\mathbf{r}') h(\mathbf{r} + \mathbf{r}') \rangle = \sigma_h^2 - \gamma_h(\mathbf{r}), \quad (6.22)$$

where the averaging is over all the positions $\mathbf{r}'$, and γ_h is the semi-variogram (see Chapter 5). For an isotropic surface, one naturally has $C_2(\mathbf{r}) = C_2(|\mathbf{r}|)$.

6.5.4

Surface Roughness

There is now ample experimental evidence (Brown and Scholz, 1985; Brown *et al.*, 1986; Brown, 1987a,b; Power *et al.*, 1987; Poon *et al.*, 1992; Schmittbuhl *et al.*, 1993a; Cox and Wang, 1993; Odling, 1994; Boffa *et al.*, 1998; Ponson *et al.*, 2007) that the internal surface of natural fractures in rock masses – and even induced fracture surface of heterogeneous materials (Sahimi, 2003b) – is very rough, with the roughness following self-affine fractal statistics described above. More specifically, we consider the internal surface of a fracture with a single-valued function $h(x, y)$ representing the surface height, and the coordinates (x, y) lying in the mean plane of the fracture. We assume the surface not to have any overhangs; that is, the surface is *continuously* rough. As described earlier, self-affinity of a rough surface implies that it exhibits scale invariance under rescaling with direction-dependent rescaling factors such that $x \rightarrow b_x x$, $y \rightarrow b_y y$ and $h \rightarrow b_h h$. We assume isotropy in the mean plane of the fracture, which is typically the case, and write $b_x = b_y = b$ and $b_h = b^H$ so that

$$h(x, y) = b^{-H} h(bx, by), \quad (6.23)$$

where H is the roughness or Hurst exponent. For example, Schmittbuhl *et al.* (1993a) measured the height of a granitic fault surface as a function of position along 1D profiles, and showed that the profiles exhibit self-affinity or anisotropic scale invariance, following Eq. (6.23).

There is no consensus on the value of the roughness exponent H , and it is perhaps too much to expect a universal value of H for all types of materials and fractures. Earlier studies (Sahimi, 1998, 2003b) indicated that the fracture surface of *disordered materials* is characterized by a more or less universal value of the roughness

exponent, $H \simeq 0.75 - 0.8$, with very weak dependence on the orientation considered. In fact, Bouchaud, E. *et al.* (1990) even conjectured that $H \simeq 0.8$ represents a universal value. However, more careful studies have indicated non-universality. It has been suggested that the roughness exponent may take on two distinct values depending on the type of the fractured materials. One is $H \simeq 0.8$ for such materials as glass, cement, granite, and tuff (Brown, 1989; Måløy *et al.*, 1992; Poon *et al.*, 1992; Schmittbuhl *et al.*, 1993a; Glover *et al.*, 1998; Auradou *et al.*, 2005; Matsuki *et al.*, 2006), and $H \simeq 0.5$ for sandstones (Boffa *et al.*, 1998; Ponson *et al.*, 2007), calcite (Gouze *et al.*, 2003) and sintered glass beads (Ponson *et al.*, 2006). However, the estimate, $H \simeq 0.75 - 0.8$, was also reported for sandstones (Nasseri *et al.*, 2006). As for the internal surface of the fracture of rock, a similar estimate, $H \simeq 0.85$, was reported by Schmittbuhl *et al.* (1993a) for granitic faults. Vickers *et al.* (1992), Cox and Wang (1993), Johns *et al.* (1993), and Odling (1994), however, analyzed extensive data for a variety of rock joints and reported nonuniversal values of the roughness exponent H in the range $0 < H \leq 0.85$.

There is also considerable indirect experimental evidence for roughness of the internal surface of fractures. For example, if we model a single fracture as the space between two parallel flat surfaces, then the volume flow rate q is related to the distance δ between the two plates by $q \propto \delta^3$, the so-called *cubic law*. Significant experimental deviations from the cubic law have been observed and attributed (Tsang and Witherspoon, 1981; Hakami and Larsson, 1996) to the roughness of the internal surface of fracture.

The evidence that we have described so far for the self-affine nature of roughness of a fracture surface has been obtained from laboratory experiments and characterization. Detailed data and measurements on field-scale fractures are scarce, and the best evidence that they provide is circumstantial. The data are typically obtained from fracture traces, but they indicate mostly jumps and discontinuities, rather than a continuously rough surface. Therefore, at large length scales, the surface roughness of fractures may not be self-affine. However, such length scales are truly large and, therefore, it is reasonable to assume that the internal surface of fractures is, up to a certain length scale, self-affine.

6.6

Characterization of Fracture Networks

A fracture network is characterized by the statistical distributions of six distinct properties of its constituent fractures that are the fractures' (1) length; (2) displacement; (3) aperture distribution; (4) orientation; (5) density, and (6) connectivity. Extensive experimental evidence indicate that most properties of rock fractures follow power-law statistics. Therefore, we first address the question regarding the prevalence of the power laws.

6.6.1

Fractures and Power-Law Scaling

Prior to the first application of fractal geometry to characterization of fracture networks of the FS porous media, data collection was limited mostly to the type of 1D sampling of spacing between fractures intersected along a traverse that was described in Section 6.1. The sampling was done without any regard for or knowledge of the fractures' orientation or size, and the data analysis was limited to the calculation of arithmetic averages of the properties of interest using such distributions as the log-normal or exponential, which did not provide deep insight into the properties of the fracture networks. In the early 1980s, a step in the right direction was taken by several research groups when they explored the use of semi-variograms as a method of analyzing fracture spacing along a scanline. As described in Chapter 5, semi-variograms plot the second moment of a distribution, for example, that of the number of fractures per unit length of the scanline as a function of the length of sampling increment over some range of the increment size. Doing so is similar to the box-counting method of estimating the fractal dimension of an object described in Section 6.2. In fact, such semi-variograms can be recast into a fractal analysis if we plot their logarithm versus logarithm of sample increment size. The slope of the resulting diagram would provide an estimate of the Hurst or roughness exponent H and, hence, yield an estimate of the fractal dimension of the object.

If fractal concepts are truly relevant to describing the properties of fracture networks, they would immediately imply the existence of power-law scalings for various characteristics of the networks. Therefore, the relevant and important question is, why should power-law scaling be favored during the process of fracture formation? The key point in answering this question is that power laws imply the absence of a characteristic length scale in the system, and this is what one expects for natural systems, for example, rock, in which the heterogeneities exist regardless of the size of the systems that one may inspect.

Strong evidence for the prevalence of power-law distributions in describing several important properties of fractures is also provided by large-scale computer simulation of fracture of heterogeneous materials. In such models, first proposed by Sahimi and Goddard (1986), one utilizes a disordered network, each bond of which describes the sample material on a small length scale, with fracture characteristics described by a few key parameters. Several types of forces that are typically encountered in the deformation of rock may act on the bonds, including stretching, angle-changing and torsional forces. One then deforms the network gradually by applying a specific boundary condition (for example, shear, tensile, etc.) to the network, as a result of which the individual bonds begin to break irreversibly, hence generating microcracks in the network. If the sample's deformation continues, the microcracks will grow, join each other, and form large fractures.

Computer simulations (Sahimi and Goddard, 1986; de Arcangelis *et al.*, 1989; Arbabi and Sahimi, 1990b; Lockner and Madden, 1991; Reuschlé, 1992; Sahimi and Arbabi, 1992, 1993, 1996; Sahimi *et al.*, 1993; Renshaw and Pollard, 1994; Robertson *et al.*, 1995; Davy *et al.*, 1995; Bonnet *et al.*, 2001; for a comprehensive review

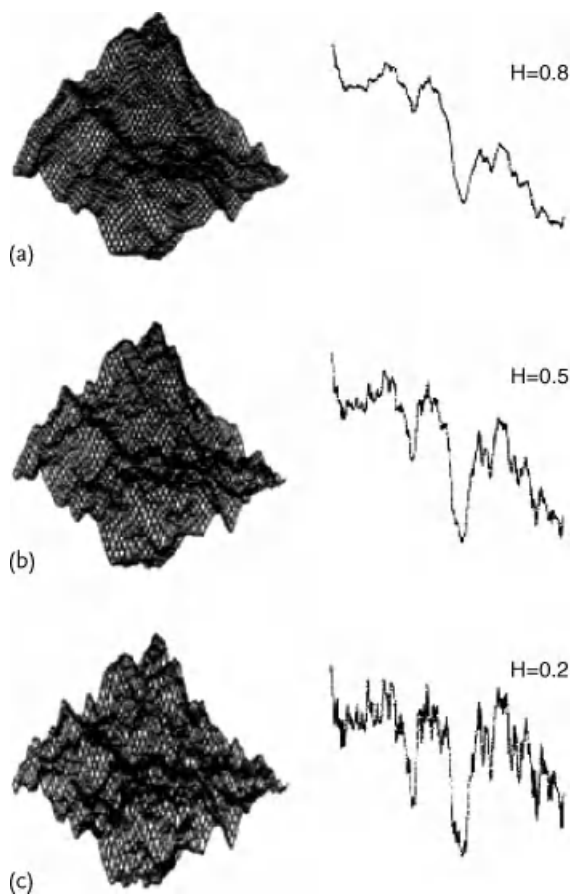


Figure 6.1 Rough interface in 1D and surface in 2D: (a) $H = 0.8$; (b) $H = 0.5$; (c) $H = 0.2$.

see Sahimi, 2003b) indicate power-law scaling for most, if not all, properties of the fracture network that is produced by such models, particularly when the disorder is broad, as is the case in rock. In addition, such models indicate power-law dependence of the strength of the rock sample on the length scale of interest, which is also predicted by rock mechanics. It is important to point out that a wide variety of models, both quasi-static and dynamic as well as a broad class of the distribution of the heterogeneities were employed in the aforementioned works and, yet, they all yielded the same type of power-law scaling laws for various properties of fracture networks, implying that such power laws may be a generic feature of the fracture of heterogeneous rock. We will come back to such models in Chapter 8, where we describe models of fracture networks.

6.6.2

Distribution of Fractures' Length

Let $n_l(\ell)$ be the number density of fractures with lengths in the interval $[\ell, \ell + d\ell]$ (for $d\ell \ll \ell$). Then, the power-law distribution of the fractures' length is defined by

$$n_l(\ell) = a_l \ell^{-a}, \quad (6.24)$$

where a_l is a constant that depends on the density of the fracture, and a is the corresponding characteristic exponent. Four types of materials have been used in laboratory experiments on fractures: Clay, plaster, rock, and sand, with the last one used with or without a basal silicon putty layer. Walmann (1998) stretched a thin layer of clay to produce a system of tensile fractures with lengths that followed an exponential distribution. Walmann suggested that a power law may result from the interaction of several exponential laws. In the experiments of Reches (1986), a transition was observed from a power-law distribution to a log-normal one as the deformation of the sample continued. Reches suggested that samples with a low density of fractures should exhibit power-law distributions, whereas the high-density rock samples should follow the log-normal distribution, although a log-normal distribution may have been caused by sample-size effects. Rossen *et al.* (2000) also reported power-law distributions in the fractured porous media that they studied.

Krantz (1983), Lockner *et al.* (1992), and Moore and Lockner (1995) analyzed the length distribution of microcracks in rock samples and found that the density of microcracks was largest close to the main fracture (which is expected, as the computer simulations mentioned above also indicate the same), and that the small cracks tend to cluster around the large fractures. Such observations agree with those made on natural faults – the largest fractures in rock – reported by Anders and Wiltschko (1994). Hatton *et al.* (1993) found a power-law distribution for the length of the microcracks with $2 \leq a \leq 2.7$, where the numerical value of the exponent a was dependent upon the fluid content of sample, with its lower values corresponding to dry samples. Velde *et al.* (1991) analyzed the length distribution of fractures during brittle-to-ductile transition using fractal (power law) concepts.

Data on the distribution of fractures' length have been obtained using 1D, 2D, and 3D samples. For all the 1D data reported, which consisted of the trace lengths of fractures that intersect a scanline (see, for example, Cruden, 1977; Priest and Hudson, 1981), the length distribution was exponential. Due to its obvious difficulties, obtaining 3D data has been very difficult. Instead, most of the reported data are from 2D maps and consist of fracture traces in a plane that intersect a 3D fractured sample. Such maps have been obtained from field and seismic data, and analysis of satellite image. In most cases, the distribution of fractures' length was reported to be of power-law type, although exponential and log-normal distributions were also reported.

It has been proposed that the exponential distribution of the fractures' length is valid when stratification of rock plays a prominent role in restricting joint growth, whereas a power-law distribution may be representative of massive rock samples

without significant mechanical effect of stratification. Bonnet *et al.* (2001) compiled the existing data and the corresponding exponents. Most of the data indicated that $a \geq 2$, although smaller values have also been reported. In addition, the data indicated that the pre-factor a_ℓ in Eq. (6.24) is *independent* of the length scale and the resolution of the observations. We must keep in mind that there are many pitfalls in estimating the exponent a and, therefore, one must be careful in analyzing any data and drawing any definitive conclusions from them.

Computer simulations of fracture growth have suggested that the distribution of fractures' length may evolve as the microcracks continue to grow. For example, Cowie *et al.* (1993) reported that the distribution of fractures' length follows an exponential law during the initial steps of the deformation of the sample when the microcracks are nucleated essentially at random. However, as the microcracks grow and begin interacting with each other, the results of Cowie *et al.* (1993) indicated a crossover to a power-law distribution of the fractures' length. Cowie *et al.* (1993, 1995) and Cladouhos and Marret (1996) reported that, in their computer simulations, the exponent a depended on the extent of the deformation, hence indicating that at long times, the growth and connection of the fractures dominate the deformation process and, therefore, with increasing time, there are fewer small fractures. Sornette and Davy (1991) investigated a model of fault growth and the possibility of a universal fault length distribution. We should point out that nucleation and propagation of fractures is a highly nonlinear process and, similar to any such process, is sensitive to the linear size of the sample. Therefore, any computer simulation of nucleation and propagation of fractures in rock samples must investigate the effect of the sample size on the results.

6.6.3

Distribution of Fractures' Displacement

Another widely studied property of fractures is the distribution of the displacement (throw) on faults (see Figure 6.2). The distribution is estimated by the 1D and 2D sampling methods (see Section 6.1). In the 1D sampling, one records the displacement of a fault where it intersects a scanline, which may not necessarily represent the maximum displacement for the fault as the displacement varies along the fault. Since the largest faults have the highest probability of intersecting the scanline, the method necessarily samples such faults. If $n_d(u)$ is the number density of the fractures that have a displacement u , the data have been fitted to a power law,

$$n_d(u) = a_d u^{-\zeta}, \quad (6.25)$$

where the reported estimates of exponent ζ obtained from the 1D measurements are in the range, $\zeta(d=1) = 0.4 - 1.0$ (see, for example, Gillespie *et al.*, 1993; Nicol *et al.*, 1996; Steen and Andresen, 1999).

If the maximum displacement $u_{\max}$ along each fault is measured, then the sampling is 2D. To carry out such measurements, one must first identify each fault. Because sampling is most often done in 2D sections from 3D samples, identification of the fault is somewhat subjective and, therefore, the measured data will not

necessarily represent the maximum displacements. Despite this fact, it is generally believed that the 2D method yields reliable data. The reported data (see, for example, Villemin and Sunwoo, 1987; Childs *et al.*, 1990; Scholz and Cowie, 1990; Gauthier and Lake, 1993; Carter and Winter, 1995; Watterson *et al.*, 1996; Yielding *et al.*, 1996; Pickering *et al.*, 1997; Fossen and Hesthammer, 1997) indicate an exponent in the range, $\zeta(d=2) = 1.7\text{--}2.4$, with an average of about 2.2, where $\zeta(d=2)$ is defined by Eq. (6.25) in which u has been replaced by $u_{\max}$. Hence, there appears to be no simple relation between the exponents obtained from the 1D and 2D sampling. The relation that is often used, namely, $\zeta(d=2) = \zeta(d=1) + 1$, assumes that the displacements and positions are independent, which is not necessarily true.

The lengths and maximum displacements of faults are generally correlated. If we write

$$u_{\max} = b\ell^\chi, \quad (6.26)$$

then, it is clear that $a = \zeta\chi$. Although theoretical considerations based on the theory of elasticity predict that $\chi = 1$, the experimental data (see, for example, Gillespie *et al.*, 1992; Fossen and Hesthammer, 1997) indicate that $0.5 \leq \chi \leq 2$. Aside from attributing the deviation of χ from unity to the scatter in the data, it has been suggested that χ is affected by faults intersecting one another, and that linkage of fault segments may result in $\chi < 1$ (see, for example, Peacock and Sanderson, 1991; Trugdill and Cartwright, 1994; Cartwright *et al.*, 1995). Other factors, for example, finite sizes of the samples and their heterogeneity, may also affect the numerical value of χ (Gross *et al.*, 1997).

6.6.4

Distribution of Fractures' Apertures

It is, of course, clear that a fracture's aperture is a crucial parameter that characterizes its capacity for fluid flow. The aperture determines a fracture's permeability since the volume flow rate Q through a fracture is given by $Q \propto \delta^m$, where $m = 3$

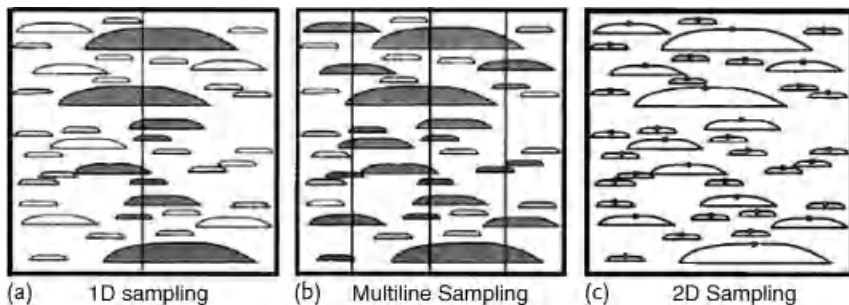


Figure 6.2 1D (a,b) and 2D sampling (c) of fault displacements. In on-line sampling (a), only displacements for those faults that intersect the line are recorded. In multiline

sampling (b), the longer faults are sampled several times. In 2D sampling (c), the maximum displacement for each fault is recorded (after Bonnet *et al.*, 2001).

for a fracture with a smooth surface. As already mentioned, however, for a fracture with rough surface, m can be as large as six. One possible source for the variation in m may be the roughness of the fracture surface. The roughness of a fracture's internal surface also implies that one must define a *local* aperture that varies spatially even within a single fracture.

Note that, as described earlier, in addition to its surface roughness, there are also voids and contact areas in a fracture that further contribute to the nonconstancy of the apertures. Moreover, apertures measured at the surface may be wider than those deep inside due to stress release. In addition, for some natural porous media, no correlation has been detected between a fracture's length and its aperture. Therefore, fractures in a network may appear to have different characteristics than if they are isolated, where the aperture is expected to correlate with a fracture's length. In any event, each fracture should, in fact, be characterized by a distribution of local apertures. Assuming that an average aperture can be unambiguously defined, a fracture network is then characterized by a distribution of *average* apertures.

The distributions of the apertures, both at the scale of a single fracture and over large length scales relevant to a fracture network, are typically broad because the apertures may change over time due to mechanical misfits of the fracture walls (see Section 6.5.1), chemical reactions, dissolution and precipitation as well as the pressure due to the depth of overburden. In addition, since fracture apertures can be measured directly in cores or outcrops, or indirectly through fluid flow measurements, the reported data may exhibit considerable scatter. As mentioned in Section 6.5.1, log-normal (see, for example, Snow, 1970; Gale, 1982, 1987; Hakami, 1995; Pyrak-Nolte *et al.*, 1997), normal (Vickers *et al.*, 1992; Hakami and Larsson, 1996; Yeo *et al.*, 1998; Matsuki *et al.*, 2006), and power-law distributions (see, for example, Barton and Zoback, 1992; Sanderson *et al.*, 1994; Belfield and Sovitch, 1995; Johnston and McCaffrey, 1996; Marret, 1996) as well as unusual distributions, for example, the Lévy-stable distribution (Painter and Paterson, 1994; Sahimi *et al.*, 1995; Painter *et al.*, 1995; Painter, 1996; Belfield, 1998), have been proposed for describing the aperture distribution. The probability density for the Lévy-stable distribution is given by (see, for example, Hughes, 1995)

$$P(x) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \cos(\omega x) \exp(-C_L \omega^\alpha) d\omega, \quad (6.27)$$

where C_L is a constant and α is called the *index* of the distribution that geometrically can be interpreted as the fractal dimension of the set of points that are visited by a random walker whose probability density follows Eq. (6.27). If $\alpha = 2$, one has a Gaussian distribution. The Lévy-stable distribution corresponds to $\alpha < 2$, in which case the second moment of the distribution is divergent.

Assuming that the apertures follow a power-law distribution,

$$n_a(\delta) = a_\delta \delta^{-z}, \quad (6.28)$$

the exponent z has been measured and reported over a broad range of apertures. For example, Barton and Zoback (1992) reported that $z \simeq 2.5$ for 1600 apertures measured in the range of millimeters to centimeters. Johnston and McCaffrey (1996) reported that $z \simeq 1.7$ – 1.8 for apertures in the range 5–50 mm, while Barton (1995b) found that $z \simeq 1.53$ for apertures in the range 1–10 mm. For smaller apertures, Belfield and Sovitch (1995) reported $z \simeq 2$ – 2.4 in the range 6–40 μm , while Wong *et al.* (1989) found, for microcracks in granite and quartzite, that $z \simeq 1.8$ for apertures in the range 0.03–10 μm . In addition, similar to fractures' displacement, attempts have been made to relate the aperture to the length of a fracture through a power-law relation (see, for example, Stone, 1984; Hatton *et al.*, 1994; Johnston, 1994; Vermilye and Scholz, 1995; Walmann *et al.*, 1996; Renshaw and Park, 1997).

6.6.5

Distribution of Fractures' Orientation

A fracture's orientation is usually quantified by its *strike* and *dip*. The strike is the trace of the intersection of a fracture with a horizontal plane. The direction of the strike is specified by its azimuth, usually counted in a degree clockwise from the north. The dip is the magnitude of the angle between a fracture and a horizontal plane, also expressed in degrees. If the strike is oriented such that the dip plunges on the right-hand side, the azimuth of the strike, which takes on a unique value between 0 and 360° , and the dip angle, which is between 0 and 90° , specify the orientation of the fracture. The distribution of the orientations is constructed based on such a definition.

6.6.6

Density of Fractures

Fracture density is an important parameter in modeling of fracture networks. The 2D fracture density ρ_2 is defined as the sum of fracture-trace lengths per unit area, while the 3D density ρ_3 is defined as the average fractured surface per unit volume of rock. If a fracture network is isotropic (which is usually not the case), then $\rho_2 = \rho_3$. One can even define a 1D fracture density ρ_1 along a straight line as the number of fracture intersections per unit length. The 3D density can be obtained from a borehole or scanline survey (see Section 6.1), with the help of a weighting of the observed fractures. However, the correct weight is important. Suppose that N fractures intersect the line, and that the i th fracture intersects it with an angle ϑ_i . Then, it is not difficult to show that the fracture density ρ_3 is given by

$$\rho_3 = \frac{1}{L} \sum_{i=1}^N \frac{1}{\sin \vartheta_i}. \quad (6.29)$$

If the angles ϑ_i are completely random, then the average value of $\sin \vartheta$ between a fracture plane and a survey line is $1/2$, and between a fracture plane and a survey plane is $\pi/4$. Thus, for this particular case, one has $\rho_1 = \frac{1}{2}\rho_3$ and $\rho_2 = \frac{1}{\pi}\rho_3$.

6.6.7

Connectivity of Fracture Networks

To characterize the connectivity and clustering of intersecting fractures (and faults), one may use percolation theory. In addition to Chapter 3, for a simple introduction to percolation theory, see Stauffer and Aharony (1994); see also Sahimi *et al.* (1990), Berkowitz and Balberg (1993), and Sahimi (1993b, 1994a, 1998, 2003a).

If fractures do not form a sample-spanning cluster between two widely-separated points, such as the injection and production wells in an oil reservoir, then the size of the clusters that they form is finite, as the network is below its percolation threshold. In this case, the typical radius ξ_p of the clusters of fracture is what we called in Chapter 3 the *correlation length*. As described there, the correlation length for the case in which the fractures do form a sample-spanning cluster – when the fracture network is above the percolation threshold – is simply the typical radius of the matrix in between the fractures. Also, recall that near p_c , $\xi_p \sim |p - p_c|^{-\nu}$, and that ν , the *critical exponent* of ξ_p , does not depend on the connectivity of the network.

The physical significance of ξ_p for fracture networks is that it sets the length scale for their macroscopic homogeneity: over *any* length scale L with $L \gg \xi_p$, the network is macroscopically homogeneous. For $\ell \ll L \ll \xi_p$, however, where ℓ is the length of a single fracture, the network is macroscopically heterogeneous. In fact, over such length scales, the sample-spanning cluster of fractures is a fractal and statistically self-similar object. That is, the average number $\langle N(L) \rangle$ of fractures within a cluster of linear size $L \ll \xi_p$ follows an equation similar to Eq. (6.3), where the averaging is taken over *all* the possible realizations (configurations) of the clusters of fractures for a fixed connectivity. Recall from Chapter 3 that at the percolation threshold p_c , the sample-spanning cluster of fractures is a fractal object at any length scale.

It has been proposed by many researchers that the connectivity of fracture network of rock may correspond to that of a percolation cluster *at*, or very close to, the percolation threshold (see, for example, Chelidze, 1982; Madden, 1983; Guéguen *et al.*, 1991; Sahimi *et al.*, 1993; Robertson *et al.*, 1995; Renshaw, 1998), although others (see, for example, Barton, 1995b) have argued that the fracture networks are well connected and, therefore, are well above their percolation threshold. Why should the structure of a fracture network of rock be similar to that of a percolation network at, or very close to, the percolation threshold? One argument that supports the hypothesis is based on what is usually referred to as the *critical path analysis* (CPA).

It was argued by Ambegaokar *et al.* (1971), and confirmed by extensive computer simulations, that transport processes, and in particular conduction, in a highly heterogeneous material can be reduced to one in a percolation system at or very near the percolation threshold. The idea is that in a broadly-disordered material, a finite volume fraction of the material possesses very small conductivity and, hence, makes negligible contribution to the overall conductivity or other effective transport properties. Therefore, zones of low conductivity may be eliminated from the material which would then reduce it to a percolation system. Ambegaokar *et*

al. (1971) described a procedure by which the equivalent percolation network is built up, and showed that the resulting model is at, or very near, its percolation threshold. The CPA has been applied to fluid flow in laboratory-scale (Katz and Thompson, 1986, 1987; Sahimi, 1993a; Friedman and Seaton, 1998) as well as FS porous media (Sahimi and Mukhopadhyay, 1996). Hunt and Ewing (2009) describe several applications of the CPA to problems in porous media. We will return to this concept in Chapters 10–12, where we describe single-phase flow and dispersion processes in porous media.

When applied to heterogeneous fractured rock, the CPA implies that the fracture network should have the connectivity of a percolation cluster because the fractures are the main pathways for fluid flow in the rock as their permeabilities (or hydraulic conductances) are much larger than those of the matrix in which they are embedded and, thus, fractured rock may be envisioned as a mixture of nearly impermeable bonds (the pores of the matrix) and the highly permeable bonds (the fractures), that is, a percolation network. Using the procedure of Ambegaokar *et al.* (1971), one then finds that the fracture network of rock must be at, or very near, its percolation threshold.

The idea that the fracture network in heterogeneous porous media should be similar to a percolation network at or very close to the percolation threshold may also be inferred from computer simulations of fracture propagation in highly disordered network, which was mentioned above. We will come back to this point in Chapter 8 where we describe models of fracture networks.

There is also considerable indirect evidence that supports such arguments. For example, Long and Billaux (1987) found that at the Fanay-Augères site in France, only about 0.1% of the fractures contributed to fluid flow at large length scales, hence indicating the existence of a barely-connected fracture network that should be similar to a percolation network at, or very near, its percolation threshold. Chelidze (1982) summarized the experimental results and showed that the percolation threshold for propagating and intersecting fractures is reached simultaneously with the development of a macrofracture that is sample-spanning.

In practice, rock fractures are distributed at all orientations and, therefore, the fracture network does not have a regular topology. Thus, the percolation threshold of a fracture network (the minimum number of intersecting fractures between two widely-separated planes) may depend on the orientation distribution of the fractures. In addition, as described above, the fractures' length typically follows a power-law distribution and, hence, the percolation threshold should depend on such a distribution (Englman *et al.*, 1983; Robinson, 1983, 1984a,b; Charlaix *et al.*, 1984, 1987a; Balberg, 1986; Balberg *et al.*, 1991; Huseby *et al.*, 1997; see also Chapter 8) since even a single long fracture might enable a fracture network to become sample-spanning. In particular, Berkowitz (1995) and Watanabe and Takahashi (1995), who studied 2D networks, found the percolation threshold p_c to depend on the second moment of the length distribution.

With 3D orthogonal families of square fracture of unit length, Robinson (1984b) estimated that $p_c \simeq 0.19$. He defined p_c as

$$p_c = \frac{N_c r^3}{L^3}, \quad (6.30)$$

where N_c is the number of fractures at the percolation threshold, r is the half-width of the fractures, and L is the linear size of the sample. If the orientations of the fractures are uniformly distributed, then Robinson (1984b) found that $0.15 < p_c < 0.3$. If the fractures are polydisperse, then p_c is defined as (Charlaix *et al.*, 1984)

$$p_c = \frac{N_c \langle r^2 \rangle \langle r \rangle}{L^3}. \quad (6.31)$$

In proposing Eq. (6.31), Charlaix *et al.* (1984) suggested that one must take into account the effect of the *excluded volume*, defined as the average volume surrounding an object representing a fracture into which the center of another object cannot lie without intersecting it. On the other hand, numerical simulations (Balberg *et al.*, 1984b) suggested that one must define p_c as

$$p_c = \frac{N_c \langle r^3 \rangle}{L^3}, \quad (6.32)$$

which is an intuitive generalization of Eq. (6.31).

Charlaix *et al.* (1984) suggested that *at* the percolation threshold of a 3D fracture network that consists of equal flat disks of radius r with a density of ρ_3 of disks per unit volume, one must have

$$\rho_3 r^3 \sim 0.15-0.3. \quad (6.33)$$

Note that r^3 essentially represents the volume of a disk. As Eq. (6.31) suggests, for polydisperse disks with a distribution of disks' radii, one must replace r^3 with $\pi^2 \langle r^2 \rangle \langle r \rangle$ in Eq. (6.30), which is nothing but $1/2 \langle \text{surface} \rangle \langle \text{perimeter} \rangle$, where $\langle \dots \rangle$ represents an average with respect to the distribution of the radii. We will come back to this important point in Chapter 8 where we describe models of fracture networks.

The above predictions were confirmed by Charlaix (1986) using Monte Carlo simulations. Robinson (1984b) found that for 3D networks of planar fractures, the percolation threshold, that is, the number of intersections per plane, is about two, whereas for 2D networks of fractures with constant length, the percolation threshold, that is, the average number of fractures intersecting a given fracture, was found to be about 3.1. Madden (1983) represented a fracture network by a tessellation of cubes and studied the connectivity properties of the network using the renormalization group theory (for the principles of renormalization group theory see Chapter 10).

Equations (6.30)–(6.32) were proposed based on the assumption that all the fractures have the same length. However, as discussed above, the distribution of the fractures' lengths is typically of the power-law type and, therefore, the length of

the fractures can significantly affect the location of the percolation threshold of a fracture network. Hence, one must define a more general relation for the fracture connectivity. We will come back to this point in Chapter 8.

6.6.8

Self-Similar Structure of Fracture Networks

In addition to the various properties of fracture networks for which power-law distributions and correlations have been exhibited, many investigators have studied the self-similar fractal structure of fracture networks. Such studies began in 1985 by the United States Geological Survey as part of the program to characterize the geologic and hydrologic framework at Yucca Mountain in Nevada (Barton and Larsen, 1985; Barton *et al.*, 1987; summarized in Barton and Hsieh, 1989). The site has been approved by the United States Department of Energy as an underground repository for high-level radioactive waste (the decision was still being challenged in courts at the time of writing this book). Barton and Larsen (1985) developed the *pavement method* of clearing a subplanar surface and mapping the fracture surface in order to measure its connectivity, trace length, density, and fractal scaling in addition to the orientation, surface roughness, and aperture.

The most significant observation of the Yucca Mountain study was that the fractured pavements had a fractal geometry and were scale-invariant, and that it was possible to represent the distribution of fractures ranging from 20 cm to 20 m by a single parameter, the fractal dimension D_f , calculated by the box-counting method, Eq. (6.1) (in which $N(r)$ should be taken to be the number of fractures of length r). For the fracture surfaces analyzed by Barton and co-workers, $D_f \simeq 1.6\text{--}1.7$. LaPointe (1988) carried out a careful reanalysis of three fracture-trace maps of Barton and Larsen (1985) and found that for the corresponding 3D fracture networks, $D_f \simeq 2.37, 2.52$, and 2.68 with an average of about $D_f \simeq 2.52$ (so that the average fractal dimension of 3D the fracture networks is essentially identical with that of 3D percolation network at the percolation threshold p_c ; see Chapter 3).

Velde *et al.* (1991) analyzed fractal patterns of fractures in granites. Vignes-Adler *et al.* (1991) carried out fractal analysis of fracturing in two African regions. They analyzed data at several length scales, ranging from those obtained from satellite images to ground states, and found strong evidence for the fractality of the fracture patterns, while 2D maps of fracture traces spanning nearly *ten* orders of magnitude, ranging from microfractures in Archean Albites to large fractures in South Atlantic seafloors, were analyzed by Barton (1992) who reported that $D_f \simeq 1.3\text{--}1.7$. The reader should also consult Walsh and Watterson (1993) on the pitfalls of fractal analysis of fracture patterns.

A similar result was obtained for the Geysers geothermal field in northeast California (Sahimi *et al.*, 1993), one of the most significant geothermal fields in the world. An example of its fracture surface is shown in Figure 6.3 (Sahimi, 1992b). The reservoir's fractures can be detected during drilling as they produce a sudden and measurable increase in the steam pressure. Analysis of the 2D data indicated a fractal dimension $D_f \simeq 1.9$ (which is the same as that of 2D percolation networks

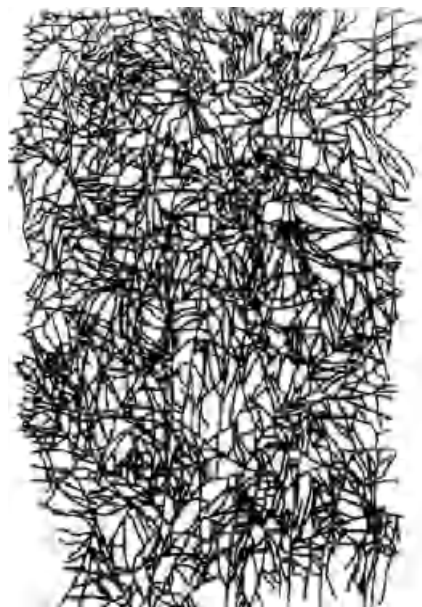


Figure 6.3 Fracture surface of the Geysers geothermal field in northeast California (after Sahimi, 1992b).

at p_c). Using a discrete model of mechanical fracture of solids developed by Sahimi and Goddard (1986), Sahimi *et al.* (1993) proposed that at *large* length scales, the fracture network of the Geysers field must have the structure of the sample-spanning percolation network at p_c .

Watanabe and Takahashi (1995) also used fractal concepts to characterize the structure of geothermal reservoirs, as did Babadagli (2001), who analyzed fractal properties of 2D samples of geothermal reservoirs in south-western Turkey. Nolen-Hoeksema and Gordon (1987) and Chelidze and Guéguen (1990) studied the fracture patterns in Stockbridge (near Canam, Connecticut) dolomite marble, which is very branched and appears to be a highly interconnected network. Analysis of Chelidze and Guéguen (1990) indicated that the 3D fracture network is a fractal object with $D_f \simeq 2.5$, very close to that of the sample-spanning percolation cluster at p_c .

In addition to fractures, faults, which can be thought of as fractures at the largest length scales, may also possess fractal properties. Faults are usually created when two strata or layers move with respect to each other. The interface between the two displaced layers is what constitutes a fault. Therefore, in some sense, faults are similar to fractures and one often finds large faults in almost any kind of reservoir. Unlike fractures, however, which are created by a variety of processes, ranging from the diagenetic to mechanical processes, faults are usually manifestations of tectonic processes that rock experienced in the past. Moreover, unlike fractures that provide large permeability zones and facilitate transport of fluids in rock, faults may or may

not do so. Faults sometimes hinder fluid transport in rock by compartmentalizing and isolating large portions of it.

Tchalenko (1970) observed that over many orders of magnitude in length scale, ranging from millimeters to hundreds of meters, shear deformation zones in rock are similar, thus suggesting strongly that the fault patterns are fractal (although Tchalenko was not aware of the fractal geometry). The work of others (Andrews, 1980; Aki, 1981; King, 1984) confirmed this suggestion. In particular, Okubo and Aki (1987) and Aviles *et al.* (1987) analyzed maps of the San Andreas fault system in California and obtained fractal dimensions for fault surfaces varying from 1.1 to 1.4 (see also Davy, 1993; Barton, 1995a; Berkowitz and Hadad, 1997) reanalyzed the data for Yucca Mountain and reported that $D_f \simeq 1.7\text{--}1.98$, which are close to the fractal dimension of 2D percolation networks at p_c .

In many cases, fracture networks are products of fragmentation processes. Rock is fragmented by joints and weathering. Explosives are often used to fragment rock. Another mechanism for fragmenting a porous formation is dissolving it in a reactant (e.g., an acid). Large fractures can form in all the cases. If the fragmentation process can give rise to a fractal fragment size distribution, then the fractures that are formed by that process may also be expected to be fractal objects, which has been found to be so in many cases. Turcotte (1986) analyzed the size distribution of rock samples that had been impacted by an explosion, and also basalt rock that had been impacted by polycarbonate projectiles and fragmented, and many other systems. He showed that the number n_f of fragments of mass M scales with M as

$$n_f \sim M^{-\tau}, \quad (6.34)$$

where τ was found to be about 0.85. Since 3τ may be interpreted as a fractal dimension, Turcotte's findings imply a fractal dimension of about 2.5. Likewise, Poulton *et al.* (1990) found that the length and spacing of discontinuities in rock masses also follow power laws, typical of fractal systems. Finally, Sahimi (1991) showed how fractal fragment size distribution and fracture patterns can arise as a result of the consumption of a porous medium by a reactant.

Before closing this section, we should point out that another way of generating porous media that resemble fractal systems is by constructal theory (see, for example, Bejan and Errera, 1997; Bejan *et al.*, 1998; Bejan and Tondeur, 1998). In this theory, the channels, locations, tributaries, and cutoffs are results of a geometric optimization process, subject to local and global constraints. Examples include volume-point flows in fractured porous media and river drainage networks. In the past, there has been ample speculations as to why fractal structures seem to be abundant in nature. Constructal theory may explain this abundance as nature's way of optimizing the structures of such systems.

6.6.9

Interdimensional Relations

An important question is how to extrapolate data for fracture networks from 1D to 2D, and from 2D to 3D. The mapping must be based on stereological methods. De-

spite the fundamental importance of the subject, few thorough studies have been carried out to investigate it. In particular, Berkowitz and Adler (1998) invoked the usual, but highly idealistic, assumption that fractures can be modeled as disks of finite radii, and predicted the distribution of fracture trace lengths on planes that intersect randomly distributed disk-like fractures. They also considered the extrapolation problem from 1D to 2D and from 2D to 3D, and developed a series of analytical relationships to accomplish their goal.

A related important question is how to relate the exponents for the various distributions, defined above, which are obtained from low-dimensional samples, to those for 3D ones. For example, how does one relate the fractal dimension of a fracture network obtained from 2D maps to 3D samples? It is common to assume that

$$D_f(d = 3) = D_f(d = 2) + 1, \quad (6.35)$$

which would be true for isotropic and large enough samples, in which the fractures are randomly distributed with a uniform spatial distribution. However, Eq. (6.35) is not necessarily valid if there are strong correlations between the spatial positions of the fractures, or if there are strong directional anisotropy. Hatton *et al.* (1993) proposed that

$$\kappa(d = 3) = c_1 \kappa(d = 2) + c_2, \quad (6.36)$$

where κ is any exponent of interest, and $c_1 = 1.28 \pm 0.3$ and $c_2 = -0.23 \pm 0.0.3$. Hatton *et al.* (1993) compared scaling exponents from acoustic emission (3D) data and direct fracture trace (2D) data in order to obtain Eq. (6.36). Borgos *et al.* (2000) studied the problem theoretically and suggested that $c_1 = 1$ and $0 \leq c_2 \leq 1$. The main conclusion is that such equations as (35) represent the ideal case and are not necessarily valid for natural rock samples that are characterized by long-range correlations, directional anisotropy, and preferential fracture nucleation at the tensile surface.

6.7

Characterization of Fractured Porous Media

In most practical situations, the FS porous media contain an extensive fracture network. If the fracture network is the main conduit for fluid flow, then the porous matrix acts only as a fluid storage area that feeds the fractures. In this case, the matrix is usually assumed to be disconnected, or impermeable. Alternatively, the matrix can also be connected and, therefore, contribute to flow of the fluids. This would then constitute the most complex type of porous media, the characterization and realistic modeling of which remain a significant challenge, despite the considerable progress that has been made over the last several decades. The characterization of fractured porous media is done in several steps. In what follows, we describe the three most important steps.

6.7.1

Analysis of Well Logs

Characterization of a fractured porous medium (or unfractured porous media for that matter) usually involves, as one of the first steps, the analysis of we call the *direct data* that are typically in the form of various logs that are collected along wells in, for example, an oil reservoir, for example, the porosity, resistivity, gamma-ray, and temperature logs. The standard method of analyzing such logs is based on the semi-variogram of the data described in Chapter 5 in order to determine the structure and nature (the probability distribution) of the data, and the extent of the correlations in the data. As pointed out in Chapter 5, in a pioneering work, Hewett (1986) proposed that the porosity logs (and, as discovered later, the permeability distributions) of most FS porous media may follow self-affine fractal distributions that were described and discussed in Chapter 5. More specifically, Hewett provided strong evidence that the porosity logs in the direction perpendicular to the bedding may follow the statistics of a fractional Gaussian noise (FGN) described in Chapter 5, whereas those parallel to the bedding follow a fractional Brownian motion (FBM), described in Chapter 5 and earlier in this chapter. The semi-variograms for FBM- and FGN-type data were given in Chapter 5. Mehrabi *et al.* (1997) showed, however, that the semi-variogram is not necessarily the most accurate and efficient method of analyzing log data.

The question then is: given a porosity log along a well (or, if possible, estimates of the permeabilities along the same well) in a FS porous medium, how can one accurately analyze it to understand its structure? In particular, if such data follow the statistics of a FBM or FGN, how can one estimate the Hurst exponent H that characterizes the correlations in the data? In the case of the FBM- and FGN-type data, it was shown by Mehrabi *et al.* (1997) that the most efficient and accurate analysis is done by either the orthonormal wavelet decomposition method, or by the maximum entropy method, both of which were described in Chapter 5 and, thus, need not to be discussed further. We should, however, point out that, while the two methods are highly accurate, in terms of the required size of the data array for reliable characterization of the long-range correlations, the maximum entropy method offers a more robust tool for analyzing a given data array (Mehrabi *et al.*, 1997). It yields valuable information and accurate results even when one has a small data array, which is a great advantage for characterization of the FS porous media, since collecting a large data base for such porous media is costly and time consuming.

6.7.2

Seismic Attributes

After the well logs are analyzed and their structure understood, they are combined with a modern geostatistical method (Chapter 5) in order to generate what is usually referred to as the *geological model* of a porous medium. However, although well logs provide good vertical resolution, which is important to accurate flow simula-

tions, they only represent a small portion of a FS porous medium. Log data also represent “point” properties because they represent the properties of portions of a porous medium that are very small compared with the medium’s overall size, or even the size of the grid blocks that are used in the simulation of flow and transport).

On the other hand, seismic data are areally dense, but vertically sparse and, therefore, are complementary to well log data. Seismic data represent, however, interval-average or “block” properties of the rock. Thus, there is a *volume support difference* between the two types of data. If one uses averaged log data, instead of “point” data, then the difference between their volume support and that of seismic data is reduced to the level that the two sets of data may be integrated with good accuracy in order to produce a more accurate representation of a FS porous medium.

Several methods have been suggested for combining the well log and seismic data. For example, Zhu and Journel (1993) proposed a method whereby the well and seismic data are encoded as local prior probability distributions. These are then updated into posterior distributions during the sequential indicator simulation process (see Chapter 5), which is used to extend the well logs and other data to the interwell zones. Values of the properties of interest are selected randomly from the local posterior distributions. Somewhat similarly, Doyen and Psaila (1994) constructed a *seismic likelihood function* based on seismic-lithotype crossplot in order to modify the local probability distributions generated by the sequential indicator simulation algorithm. In contrast, Fournier and Derain (1995) performed multivariate analysis on a calibration data set that consisted of well logs and nearby seismic traces in order to establish a non-parametric correlation between the well log data and some seismic attributes. To obtain the correlation, averaged (instead of point) properties were used. The resulting correlation was then utilized with the seismic data in order to obtain seismic-derived properties which were then combined with well log data using co-kriging (Chapter 5). The method developed by Zhu and Journel may be superior to that of Fournier and Derain, at least when applied to synthetic seismic data.

Behrens *et al.* (1998) introduced an accurate method by which seismic attributes are incorporated into the 3D model of a FS porous medium, for example, an oil reservoir, using a geostatistical method that explicitly takes into account the difference in the vertical resolution of the well log and seismic data. Suppose that $Z(r_i)$ ($i = 1, \dots, n$) represent the sample well data in the search neighborhood (“point” data), and that $\langle Z(r) \rangle$ is the arithmetic average of $Z(r_i)$ in the vertical column that contains the point r_i (“block” data). One then determines the estimates $Z^*(r)$ and the block kriging variance $\sigma_{BK}^2(r)$ from

$$Z^*(r) - m = \sum_{i=1}^n \lambda_i [Z(r_i) - m] + \lambda_s [\langle Z(r) \rangle - m] , \quad (6.37)$$

$$\sigma_{BK}^2(r) = C_{PP}(0) - \sum_{i=1}^n \lambda_i C_{PP}(r, r_i) - \lambda_s C_{PB}(r, r) , \quad (6.38)$$

where λ_i , $i = 1, \dots, n$, and λ_s are the usual kriging parameters (see Chapter 5) that are determined from the following set of equations:

$$\sum_{j=1}^n \lambda_j C_{PP}(r_j, r_i) + \lambda_s C_{PB}(r, r_i) = C_{PP}(r, r_i), \quad i = 1, \dots, n, \quad (6.39)$$

$$\sum_{j=1}^n \lambda_j C_{PB}(r, r_j) + \lambda_s C_{BB}(r, r) = C_{PB}(r, r). \quad (6.40)$$

Here, $m = E\{Z(r)\} = E\{\langle Z(r) \rangle\}$ is the stationary mean, $C_{PP}(r_i, r_j)$ is the point-to-point covariance between the locations r_i and r_j , $C_{PB}(r, r_i)$ is the point-to-block covariance between point r_i and the vertical column that contains r (that is, the average point-to-point covariance between r_i and all the points within the column containing r), and $C_{BB}(r, r)$ is the block-to-block covariance of the column that contains r with itself (that is, the average point-to-point covariance between pairs of points within the column that contains r ; see, for example, Isaaks and Srivastava, 1989, for more details).

To implement the method, the data $Z(r)$ and $\langle Z(r) \rangle$ are first transformed to normal-score data with zero mean and unit variance. Suppose that $Y(r)$ and $\langle Y(r) \rangle$ are the corresponding normal-score data, and that C_{PP} , C_{PB} , and C_{BB} denote the corresponding point-to-point, point-to-block, and block-to-block correlogram model. Clearly, by definition, $E\{Y(r)\} = E\{\langle Y(r) \rangle\} = 0$, $\sigma^2\{Y(r)\} = 1$, and $\sigma^2\{\langle Y(r) \rangle\} = \sigma^2\{Y(r)\} - [1 - C_{BB}(r, r)] = C_{BB}(r, r)$. Therefore, Eqs. (6.37) and (6.38) are rewritten as

$$Y^*(r) = \sum_{i=1}^n \lambda_i Y(r_i) + \lambda_s \langle Y(r) \rangle, \quad (6.41)$$

$$\sigma_{BK}^2(r) = 1 - \sum_{i=1}^n \lambda_i C_{PP}(r, r_i) - \lambda_s C_{PB}(r, r), \quad (6.42)$$

and the kriging parameters are the solutions of the following equations,

$$\sum_{j=1}^n \lambda_j C_{PP}(r_j, r_i) + \lambda_s C_{PB}(r, r_i) = C_{PP}(r, r_i), \quad i = 1, \dots, n, \quad (6.43)$$

$$\sum_{j=1}^n \lambda_j C_{PB}(r, r_j) + \lambda_s C_{BB}(r, r) = C_{PB}(r, r). \quad (6.44)$$

Therefore, the entire procedure may be summarized as follows:

1. We first transform the original well data to normal-score data with zero mean and unit variance. Likewise, the original seismic-derived map data are transformed to normal-score data with zero mean and variance $C_{BB}(r, r)$.
2. A random path that visits all the points to be simulated is defined.
3. At each node in the path, the Gaussian conditional density function with the mean and variance that are given by Eqs. (6.41) and (6.42) is constructed using

all the original normal-score data and all the previously simulated values that are within the search neighborhood.

4. A random number is generated according to the Gaussian distribution constructed in item 3.
5. We return to item 3 and repeat the procedure until all the nodes have been visited.
6. We then back-transform the final realization of the FS porous medium into the original space.
7. We must also honor seismic precision by calculating a scale factor that uniformly tightens or expands the plot of input seismic map versus column averages of the resulting model.

Behrens *et al.* (1998) showed that this method results in very accurate models of several oil reservoirs that they studied.

Let us mention that in recent years, increasing use has been made of 4D seismic reservoir monitoring and modeling. By 4D, we mean time-lapse 3D seismic, or repeated 3D seismic surveys over a period of time. Such 4D data have indicated significant potential for mapping out the distribution of fluids in the interwell zones, hence helping one to monitor oil reservoirs, planning for their management, monitoring fluid contacts, characterizing the flow properties of faults, and identifying pressure compartmentalization. Lumley and Behrens (1998) present a detailed discussion of the possible problems that one might encounter in utilizing 4D seismic data and the solutions to such problems.

6.7.3

Fracture Distribution

The next step of characterization of a fractured porous medium involves identification of the spatial distribution of the fracture to be included in the model of the medium. Methods for characterization of fractures and fracture networks that were described in Sections 6.5 and 6.6 may be used for this purpose. In addition, other types of data may provide insight into the spatial distribution of fractures in a geological formation, including oil reservoirs and groundwater aquifers. For example, mud loss data that are usually recorded during drilling of the wells are usually utilized to obtain information about the locations at which fractures may intersect the wells. The mud loss usually occurs where the fractures intersect the path of the drilling.

Fracture depth, dip, orientation, and height can be determined from the wellbore image (sonic) logs. Typically, fractures terminate against bedding surfaces that are, for example, shale layers and are a few inches to a foot in height. Outcrops are used to obtain data on fracture spacing to bedding thickness. Such data are then combined with the wellbore data. Narr (1996) proposed an accurate method for estimating fracture spacing. Some data are obtained on fracture density along the wellbores, and geostatistical methods are then used to interpolate the data to the interwell zones. The length of fractures are inferred from analog outcrops. Although,

in some cases, one may not have any correlation between the fracture aperture and length, though, in many cases, such correlations do exist. Fracture apertures can also be estimated from flow tests on core data for several wells.

Three-dimensional seismic data also provide key information on the distribution of the faults, which can be thought of as the largest-scale fractures (notwithstanding the detrimental effect that they sometimes have on fluid flow). In addition, a seismic fault map can be used to define fault control on small-scale fracturing. Often, fractures tend to be more densely populated near subseismic scale faults. The fault map is typically utilized for controlling the structural map of a FS porous medium, for example, an oil reservoir. It is also used sometimes to modify the transmissibilities in the flow simulation grid. In addition, attributes related to the fault network can be computed to map out the faults' areas of influence on the properties of the reservoir.

6.7.4

Fracture Density from Well Log Data

In case sonic logs are not available, Sahimi and Hashemi (2001) proposed a method for identifying the distribution of fractures along wells. Their method is based on wavelet transformation (WT) of the porosity (or resistivity) logs, which are usually available for any oil reservoir. In Chapter 5, we described how the WT may be used in analyzing wellbore logs. We now describe in detail how they are utilized for identifying the fracture distribution and density along the wells. The wavelet transformation of the distribution of porosity $\phi(x)$, defined by

$$\hat{\phi}(a, b) = \int_{-\infty}^{\infty} \phi(x) \psi_{ab}(x) dx, \quad (6.45)$$

is its representation in the wavelet space at different length scales and positions x using wavelets that are defined as a set of functions generated from a mother wavelet $\psi(x)$ by, $\psi_{ab}(x) = \psi[(x - b)/a]/\sqrt{a}$, where $a > 0$ is the rescaling parameter and b represents translation of the wavelet. The contraction/expansion/rescaling properties of a WT enable one to analyze $\phi(x)$ at increasingly finer or coarser length scales. Because the typical direct data, such as, the various wellbore logs, are discrete and 1D, one should utilize the 1D discrete WT. To construct such a WT, one sets $a = 2^j$ and $b = 2^j k$ in Eq. (6.45), where j and k are both integer. The resulting discrete WT of $\phi(x)$ is also called the *wavelet-detailed coefficient* (WDC) of $\phi(x)$.

The key idea is that the WDCs separate the data into different groups depending on the length scales, and have four key properties that may be exploited for identifying the distribution of fractures along wells.

1. The WDCs can be quite large even if the numerical value of the datum at x is small, and vice versa. Thus, each piece of the data is given its proper weight that, for the problem at hand, implies that, although the (total) porosity of the

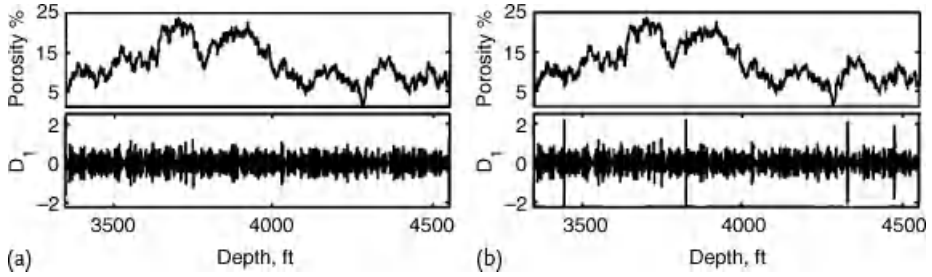


Figure 6.4 (a) Top is a porosity log that is described by a fractional Brownian motion with the Hurst exponent $H = 0.2$; its WDCs, denoted by D_1 , are shown at bottom. (b) Top is a porosity log in which some points represent

the intersections of fractures with the well along which the log was measured, with the corresponding WDCs shown at bottom, indicating the fractures' locations (after Sahimi and Hashemi, 2001).

fractures is much smaller than that of the porous matrix, its significance is properly reflected in its WDCs.

2. Local maxima in the WDCs spectrum provide clues about the scales at which important features provide significant contributions. The contributions are made either by one large feature (a single large fracture), or by a series of smaller features (several small fractures).
3. If a data array is composed of two (or more) *distinct* segments (types of data), each having their own distribution, the WDCs distinguish one from another, implying for the problem of identifying fracture along wells that because the porosity distributions of fractures and pores are different, the WT “senses” the difference between the two and, thus, the difference is reflected in their WDCs.
4. The WDCs are influenced by *local* events, which is in contrast with the spectral density of the data that is affected by the data *over the entire domain*. The implication for the present problem is clear: since the passage from the porous matrix to a fracture (and vice versa) is a local event, the presence of the fractures should be reflected in the WDCs at the correct spatial positions.

Suppose that a discrete set of data has been collected for an unfractured zone of a porous medium. Then, the plot of the data's WDCs versus the data's locations at which they had been collected (e.g., the depths along the wells) is a featureless plot in the sense that the WDCs fluctuate around a well-defined mean, exhibiting no particular features because all the data have the same distribution (property 2 above). An example is shown in Figure 6.4. If the data are, however, for a fractured zone, then the plot of the WDCs versus their locations exhibits local maxima (properties 1 and 4 above) where the fractures intersect the wells. An example is also shown in Figure 6.4. This is the essence of the method proposed by Sahimi and Hashemi (2001).

If the fractures are of the mechanical (as opposed to diagenetic) type, then the method proposed by Sahimi and Hashemi (2001) can be used for obtaining very useful information on the fracture distribution and density in the interwell zones.

Modern geostatistical approaches honor the data in models of the FS porous media, such as, an oil reservoir. For the interwell zones for which no data are available, the geostatistical methods provide estimates of the properties by an accurate method called the random residual additions (see Chapter 5) by which one adds stochastic uncertainties to the deterministic interpolated values of the properties for the unsampled zones. When combined with the seismic attributes described above, the method usually provides an accurate porosity distribution for the entire porous medium. One can then analyze the map by the method proposed by Sahimi and Hashemi (2001) in order to determine the spatial distribution of the fractures and the associated uncertainty in it.

7

Models of Porous Media

Introduction

In Chapters 4–6, we learned how various properties of porous media – fractured and unfractured – are measured, analyzed and interpreted. The natural question that comes to mind is: how do we model porous media? Any realistic modeling of flow and transport phenomena in a disordered porous medium must include, as the first ingredient, a realistic model of the porous medium itself. However, any model that we may use should depend on the type of the porous media that we wish to study and compute its various properties as well as the computational limitations that we may have. In fact, while many fundamental models for natural porous media, for example sandstones and carbonate rock, have been developed, the present computational capabilities do not allow their routine use in the simulation of complex phenomena, such as, the immiscible and miscible displacement processes. Thus, we still must utilize models that are simple enough for use in computer simulation of various flow and transport phenomena with reasonable computation, yet contain the essential features of the porous medium of interest.

In this chapter, we describe and discuss various models of unfractured porous media. Models of fractures, fracture networks, and fractured porous media will be described in Chapter 8. We consider models in which one may incorporate three fundamental scales of heterogeneities described in Chapter 1, namely, microscopic (pore level), macroscopic (core plug level) and megascopic (field-scale) heterogeneities. Porous media with the first two types of heterogeneities constitute what we refer to as the laboratory-scale porous media, while those with the third type of heterogeneities represent large-scale porous media, for example, oil reservoirs and groundwater aquifers.

7.1

Models of Porous Media

Pore space models are needed for estimating the flow and transport properties and other important dynamical features of porous media. The simplest of such prop-

erties are perhaps the effective permeability K_e and electrical conductivity g_e of a fluid-saturated porous medium. The simplest property of a pore space is its porosity ϕ and, therefore, an obvious goal for many years was to find a relationship between ϕ and K_e , the existence of which had seemed so obvious that in the early literature on flow of oil through reservoir rock, no distinction had been made between K_e and ϕ ; it had been assumed that K_e and ϕ are proportional. Later on, many empirical correlations between K_e and ϕ were suggested, the best-known of which was perhaps that due to Rose (1945) who proposed that

$$K_e \sim \phi^{m'} \quad (7.1)$$

where m' is some undetermined constant. Equation (7.1) is similar to the well-known Archie's law (Archie, 1942) for the electrical conductivity of a fluid-saturated porous medium given by

$$g_e = g_f \phi^m, \quad (7.2)$$

where g_f is the fluid's conductivity. In general though, there *cannot* be any general and *exact* relationship between K_e and ϕ because, obviously, two porous media that have the same porosity may have very different effective permeabilities since how the porosity is distributed in a material is crucial to its effective properties. Such an obvious example prompted Cloud (1941) to conclude that *there is no sensible relation between porosity and permeability*.

Over the years, many models of porous media have been developed, most of which have been motivated by a certain phenomenon. However, often while a model could be used to study a particular phenomenon and predict some of its properties, it was not general enough to be useful for studying other types of phenomena. In addition, such models often contained parameters that were either defined very vaguely, or had no physical meaning whatsoever. The sole purpose of such parameters was to make the models' predictions agree with the experimental data for a particular phenomenon.

7.1.1

One-Dimensional Models

In this class of models, the pore space is envisioned to be made of a bundle of parallel capillary tubes (pores), or a collection of tubes in series. The radius of the tubes can be the same for all, or it can be selected from a pore size distribution. The tubes can be all cylindrical, or may have converging-diverging segments. Sometimes, the tubes are deformed in order to give them a local tortuosity, or they may be given periodic constrictions. None of such models can take into account the effect of the interconnectivity of the pores, the existence of closed loops of interconnected pores, and so on. As a result, many predictions of such models are grossly in error. Scheidegger (1974) and van Brakel (1975) give lucid discussions of such models.

7.1.2

Spatially-Periodic Models

The spatially-periodic models of porous media models have been described by Nitsche and Brenner (1989) and Adler (1992). In this class of models, the pore space is represented by a periodic structure, the unit cell of which can be a capillary periodic network or some other geometrical element. An example is shown in Figure 7.1. A spatially-periodic model is characterized by an associated lattice that contains the translational symmetries of the porous medium for which the model is intended. Due to its periodic structure, the lattice is of infinite extent and is generated from any one lattice point by discrete displacements of the form

$$\mathbf{R} = i_1 \mathbf{e}_1 + i_2 \mathbf{e}_2 + i_3 \mathbf{e}_3, \quad (7.3)$$

where $\mathbf{I} = (i_1, i_2, i_3)$ is a triplet of integers, and $\{\mathbf{e}_1, \mathbf{e}_2, \mathbf{e}_3\}$ is a triad of the basic lattice vectors. The triad is *not* unique because by applying any unimodular 3×3 matrix with integer entries to the basis $\{\mathbf{e}_1, \mathbf{e}_2, \mathbf{e}_3\}$, one obtains another equally valid basis. It is often convenient to use cells that are parallelepipeds, that is, built on a given choice of basic lattice vectors. Given the flexibility that one is afforded with the choice of the unit cells, their shape is often ambiguously defined.

Spatially-periodic models are also characterized by two length scales. One is the microscopic length scale l_m of the lattice, defined as, $l_m = \max[d_{\min}(\mathbf{r})]$, where $d_{\min}(\mathbf{r})$ is the distance between the point at $\mathbf{r}$ and the nearest lattice points. For example, for a cubic lattice of lattice constant a , $l_m = \sqrt{3}a/2$. The second length scale L is one over which the averages of the physical fields of interest, for example, the pressure or concentration field, vary in a reasonable (relatively smooth) manner. This length scale is typically of the order of the linear size of the porous medium for which the model is intended. For a porous medium to be macroscopically homogeneous, one must have $L \gg l_m$.

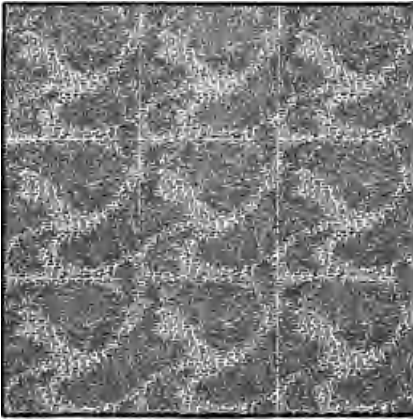


Figure 7.1 A spatially-periodic model of disordered porous media.

The simplest spatially-periodic lattice model consists of a two-dimensional (2D) array of circular (infinitely-long) cylinders which represents a relatively simple model of unconsolidated porous media if the cylinders are not allowed to overlap. Despite its simplicity, no rigorous results for flow and transport processes in such a model were obtained until Sangani and Acrivos (1982) analyzed square and hexagonal arrays of circular cylinders, determined their permeability, and discussed the application of the results to heat transfer in unconsolidated porous media. Later, Larson and Higdon (1987) considered flow in the same periodic lattices in both the axial and transverse directions. However, it was, in fact, Hasimoto (1959) who derived the first results for the effective permeability of 3D periodic lattices of spheres – a reasonable model of unconsolidated porous media and packings of particles – in the limit of small volume fraction of the spheres. The first set of results for the full range of the spheres' volume fraction was derived by Zick and Homsy (1982) and Sangani and Acrivos (1982). Their results will be described in Chapter 9.

The analysis of flow and transport processes in such models is a relatively simple problem when numerical or analytical calculations are confined to a unit cell. In principle, the unit cell may have an arbitrary shape, but if one is to analyze a *disordered* unit cell (see Figure 7.1) of arbitrary shape, the analysis would be no easier than that of non-periodic models of porous media described below. In a sense, spatially-periodic models represent a type of mean-field approximation to the truly disordered media because they do not contain any real heterogeneities and attempt to mimic the properties of the disordered media in some average way.

In some cases, the predicted effective properties do come close to those of some real disordered media. In fact, over 50 years ago, Philip (1957) stated that, “[the] particular case of flow through a cubical lattice of uniform spheres ... appears capable of providing information on permeability-geometry relations.” This statement turned out to be true in the case of the models studied by Hasimoto (1959), Zick and Homsy (1982), and Sangani and Acrivos (1982). Brenner (1980), Carbonell and Whitaker (1983), and Eidsath *et al.* (1983) studied hydrodynamic dispersion in spatially-periodic models (see Chapter 11), and found a qualitative agreement between some of their results and the experimental data of Gunn and Pryce (1969). Ryan *et al.* (1980) showed that the predicted effective reaction rate of a spatially-periodic model provides a useful estimate for some highly unconsolidated porous media, such as, packed beds. We should, however, point out that the main reason for the agreement between the predictions and the experimental data in all of such studies is that the geometry of the models closely resembled that of the experimental system. For example, Gunn and Pryce (1969) performed their dispersion experiments in a spatially-periodic porous medium.

Nitsche and Brenner (1989, p. 244) argued that, “[while] any given model of sample porous rock cannot generally be expected to possess perfect geometrical order, this does not mean that a spatially-periodic model is not useful for understanding the fundamentals of a penetrant fluid flow through its interstices.” Nonetheless, the usefulness of such models for predicting the effective flow and transport properties of, as well as numerical simulation of various phenomena in disordered and

consolidated porous media, is very limited. Nitsche and Brenner (1989) and Adler (1992) provide an extensive list of references for spatially-periodic models.

What are the main shortcomings of spatially-periodic models of porous media?

1. Regular arrays of spheres (or other particles or inclusions with regular shapes) are limited to relatively low maximum volume fractions of the spheres which are significantly *below* the solid (matrix) volume fraction of many real porous media.
2. Flow in regular lattices of isolated spheres occurs *around* the spheres, instead of *through* the narrow pores found in real porous media.
3. The spatially-periodic models may be useful for *unconsolidated* porous media in which the solid phase does *not* form a sample-spanning percolation cluster, whereas in consolidated porous media, for example, sandstone, both the solid and the fluid phases are macroscopically connected.

While the effect of solid volume fraction may not be very important for estimating the single-phase permeability of a porous medium if the heterogeneities are not broadly distributed, it is important to other flow phenomena in porous media, such as, two-phase flow, hydrodynamic dispersion, and so on, and even to single-phase flow if the medium is highly disordered. Moreover, for heat transfer in porous media (e.g., geothermal reservoirs), the effect of heat conduction through the solid matrix is important and, clearly, heat conduction through a sample-spanning solid matrix is completely different from that in isolated solid inclusions.

To extend the applicability of the spatially-periodic models to consolidated porous media, Larson and Higdon (1989) developed an interesting extension. Beginning with a regular (spatially-periodic) lattice of spheres, they allowed the sphere radii to increase beyond the point of touching in order to form overlapping spheres. Clearly, the solid fraction in this model can be anywhere between the original fraction before the growth of the spheres was started, and unity. Different lattices result in different pore shapes and sizes. Such a model is similar to the grain consolidation model of Roberts and Schwartz (1985) described below, except that Roberts and Schwartz mostly used a *random* distribution of spheres, whereas Larson and Higdon used only a spatially-periodic lattice as the starting point. The advantage of the model by Larson and Higdon is that it is amenable to certain analytical and semianalytical calculations and, at the same time, mimics certain features of consolidated porous media.

7.1.3

Bethe Lattice Models

Next to spatially-periodic models are branching network models. These are nothing but the Bethe lattices of a given coordination number Z (see Chapter 3) that have been used routinely in the statistical mechanics literature for investigating critical phenomena in the mean-field approximation. As far as their applicability to

modeling porous media is concerned, branching networks suffer from two major shortcomings.

First, although they contain interconnected bonds (pores) that may mimic that in a pore space, they lack closed loops of bonds, which are a major element of the topology of any real pore space, particularly natural porous media.

Second, for a Bethe lattice of coordination number Z , the ratio of the number of sites on the external surface of the network and the total number of sites is $(Z - 2)/(Z - 1)$ (Ziman, 1979), which takes on finite values for any $Z \neq 2$, whereas for large 3D networks, the ratio is very small. Thus, surface effects may strongly affect any property of a Bethe lattice and sometimes lead to anomalous phenomena, for example, those described by Hughes and Sahimi (1982) who investigated diffusion in Bethe lattices. The advantage of the Bethe lattices is that it is often possible to derive exact analytical formulae for the properties of interest, and sometimes, surprisingly, the predictions of such formulae agree well with those of 3D systems. Examples include diffusion and conduction in disordered Bethe lattices that will be described in Chapter 10.

Liao and Scheidegger (1969) and Torelli and Scheidegger (1972) were the first to use the Bethe lattices for modeling transport in porous media. They studied hydrodynamic dispersion in a porous medium modeled by a Bethe lattice of a given coordination number. In particular, Torelli and Scheidegger showed that such a model is fairly successful in predicting the dependence of the dispersion coefficient on the average flow velocity (see Chapter 11). Others have also used the Bethe lattices to model transport and reactions in porous catalysts (for a review see Sahimi *et al.*, 1990).

7.1.4

Pore Network Models

Chapter 3 provided the theoretical foundations for pore network models, and described the way they are developed. At the same time, a pore network model of porous media is intuitively appealing because it is clear that a fluid's paths in a porous medium branch out and, later on, join one another. A pore network model of a pore space also provides precise meaning to the concept of the pore size distribution. The network that results from the mapping usually has a random topology, and its coordination number varies from node to node. Thus, random networks such as the Voronoi network (see Chapter 3) have also been used in the literature as models of porous media (see, for example, Jerauld *et al.*, 1984b,d; Sahimi and Tsotsis, 1997; Rajabbeigi *et al.*, 2009a; Rajabbeigi *et al.*, 2009b).

The development of pore network models has been underway for several decades. Bjerrum and Manegold (1927) used a random network made of randomly distributed points in space connected to one another by cylindrical tubes in order to study transport in porous media. However, the computational limitations of that era severely limited their ability for carrying out any extensive computations. Extensive analytical calculations with such models were first carried out by de Josselin de Jong (1958) and Saffman (1959) in the context of hydrodynamic

dispersion in porous media that will be described in Chapter 11. Nevertheless, in order to make their analytical calculations tractable, they both had to make certain assumptions. As pointed out in Chapter 3, the first application of network models to modeling two-phase flow in porous media was pioneered by Fatt (1956a,b,c). He used various 2D networks of bonds representing the pore throats. The radius of the bonds was selected from a probability density function, representing the pore size distribution of the medium. No volume was attributed to the nodes. The length of each bond was assumed to be proportional to the inverse of its radius. We will come back to this problem in Chapter 15.

Later, Rose (1957) and Dodd and Keil (1959) used pore network models to study immiscible displacement processes in porous media. We already mentioned in Chapter 4 the work of Ksenzhek (1963) who used a pore network model to predict the capillary pressure curves for porous media. Thus, although in the physics literature two seminal papers of Kirkpatrick (1971, 1973) are generally credited for popularizing the use of networks of interconnected bonds for modeling transport in disordered media, the earlier pioneering works had already used such models to study transport processes in disordered porous media. Note that computer simulations of Jerauld *et al.* (1984b,d) showed that as long as the average coordination number of a topologically-random network is very close to, or identical with, the coordination number of a regular network, the effective flow and transport properties of the two networks are nearly equal.

The pore network models described so far are, in some sense, *mathematical models* that are used in computer simulations of flow phenomena in porous media. Another class of pore network models consists of a *physical network* – the man-made and transparent networks of pore bodies and pore throats. Such models have been developed for flow visualization studies, and have been particularly useful for gaining a deeper understanding of the displacement of one fluid by another. The first of such models – usually referred to as micromodels – was constructed by Chatenever and Calhoun (1952), who made bead packs from single layers of glass and Lucite beads, and studied the displacement of oil with brine. Mattax and Kyte (1961) made the first etched glass network to study displacement processes in porous media, and Davis and Jones (1968) significantly improved their technique by introducing photoetching techniques. Bonnet and Lenormand (1977) developed a resin technique for controlling the geometry of the network. Currently, etched glass and molded resin are routinely used for constructing most micromodels. Lenormand (1990) and Buckley (1991) reviewed various techniques of constructing such physical networks.

7.2

Continuum Models

Generally speaking, there are at least three different classes of continuum models of porous media.

1. Models that are made of a distribution of inclusions, such as circles, ellipses, cylinders, spheres, or ellipsoids, in an otherwise uniform background. The inclusions may or may not overlap. If they do not, the model represents an unconsolidated porous medium. If they do overlap, the model may be a reasonable representation of consolidated porous media.
2. Model that are based on tessellating the space into regular or random polygons or polyhedra and designating at random (or based on a correlation function) some of the polygons or polyhedra as representing the fractures, with the rest representing the pore space. This class of models will be described in the next chapter, where we will describe models of fracture networks.
3. Those in which one distributes sticks of a given aspect ratio or plates of given extents. Such models have been used for representing fibrous materials – a special type of porous materials (such as printing paper) – with the sticks representing the fibers. Alternatively, the sticks or plates may be thought of as the fractures in a porous medium. We will return to such models in the next chapter.

The main attractive feature of such models is that with the appropriate choice of the parameters (to be described below), they may represent many real heterogeneous porous media. Their main disadvantage is the complexities that are involved in the study of transport processes through them. Thus, although the effective transport properties of such continuum models can be and have been computed using a variety of techniques, they are still too complex for routine use in the investigation of many important phenomena in disordered porous media.

7.2.1

Packing of Spheres

Consider a statistical distribution of N identical d -dimensional spheres of radius R , which we refer to as phase 2, distributed in an otherwise uniform background that represents phase 1 of a two-phase disordered material. The system's total volume is Ω . The model is not as simple or restricted as it may seem. For example, by allowing the particles to overlap and cluster, one can generate a wide variety of models with complex microstructures. Its 2D version, that is, a system of disks or, equivalently, a system of infinitely-long cylinders distributed in the matrix can be utilized for modeling fiber-reinforced materials and thin porous films. An example is shown in Figure 7.2, where the disks' sizes are distributed according to a uniform probability density function. The 3D version can be used for modeling unconsolidated porous media (for example, packed beds of particles) if the spheres do not overlap, and consolidated porous media if they do.

If the spheres are not allowed to overlap, then one obtains what is popularly referred to as a *fully-impenetrable* or *hard-particle* model. In addition to modeling of a packing of particles, the model has been used in the study of a variety phenomena, for example, powders, cell membranes (Cornell *et al.*, 1981), thin films (Quickenden and Tan, 1974), particulate composites, colloidal dispersions (Russel

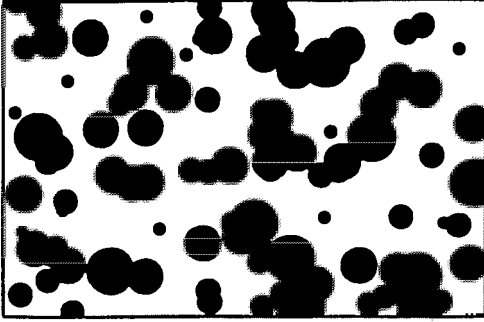


Figure 7.2 A 2D continuum model of porous media represented by a random distribution of disks in a uniform background.

et al., 1989), and granular materials such as powders. The model becomes quite general if the spheres are allowed to overlap (Weissberg, 1963). The intersection of the spheres does not have to represent a true physical entity, but can only be a way of generating a heterogeneous porous medium with a certain microstructure. An example is the *penetrable-concentric shell* model or the *cherry-pit* model (Torquato, 1984) in which each d -dimensional sphere of diameter $2R$ is composed of a hard impenetrable core of diameter $2\lambda R$, encompassed by a perfectly penetrable shell of thickness $(1 - \lambda)R$. An example is shown in Figure 7.3.

The limits $\lambda = 0$ and one correspond, respectively, to the cases of fully-penetrable sphere model, also called the Swiss-cheese model (see Chapter 3), and the totally-impenetrable sphere model – a packed bed of particles. Thus, varying λ between zero and one allows one to tune the connectivity of the particles (i.e., the number of

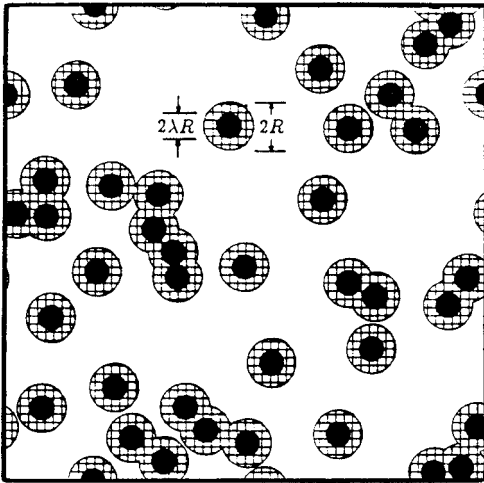


Figure 7.3 A 2D example of the cherry-pit model in which a shell of thickness $(1 - \lambda)R$ of the disks is fully penetrable (after Torquato, 1991).

particles that are in touch with a given particle) and obtain a wide variety of models with the desired morphology. In the fully-penetrable sphere model (when $\lambda = 0$), there is no correlation between the particles' positions. If ϕ_2 is the volume fraction of the particles, then the particle phase becomes sample-spanning (i.e., percolating) for $\phi_{2c} \simeq 0.67$ and 0.29 , for $d = 2$ and 3 , respectively. Note that, as pointed out in Chapter 3, no bicontinuous structure exists in 2D and, therefore, when the particle phase is sample-spanning, the pore space is not. On the other hand, for $d = 3$, the system can be bicontinuous. In particular, both the particle phase and the pores are sample spanning for $0.3 \leq \phi_2 \leq 0.97$.

A related model is the so-called *equilibrium hard-sphere* model in which the particles do not interact if the interparticle separation is larger than the sphere diameter, but there is an infinite repulsive force between them if the distance is less than or equal to the sphere diameter. An important property of this model is that the impenetrability constraint does not uniquely specify the statistical ensemble. That is, the system can be either in thermal equilibrium, or in one of the infinitely many non-equilibrium states. A fundamental difference between the equilibrium 2D hard-sphere model and its 3D counterpart should be noted. The difference is due to the fact that in 2D, the densest *global* packing is consistent with the densest *local* packing. The maximum number of d -dimensional spheres that can be packed in such a way that each sphere touches the others is $d + 1$. This implies that the coordination number of the 3D pore network that results from mapping the channels between the touching spheres onto a lattice is *exactly* four. The d -dimensional polyhedron that results by taking the spheres' centers as vertices is a *simplex* resulting in line segments, equilateral triangles, and regular tetrahedra for $d = 1, 2$, and 3 , respectively. Whereas identical simplices can fill 1D and 2D systems with overlapping, they cannot do so in 3D, that is, one cannot fill a 3D system with identical non-overlapping regular tetrahedra. Due to this inconsistency between the local packing rules and the global packing constraints, the 3D systems are said to be *geometrically frustrated*.

7.2.2

Particle Distribution and Correlation Functions

A dispersion of spheres is characterized by a number of fundamental microstructural properties, including several n -point correlation and distribution functions (Torquato, 2002; Sahimi, 2003a). The most important of such distribution, correlation functions, and microstructural properties are as follows. In the discussions below, when we speak of porous media, we mean those that are represented by the dispersion of spheres.

1. The most fundamental distribution function is $\rho_n(\mathbf{r}^n)$, where $\rho_n(\mathbf{r}^n)d\mathbf{r}_1 d\mathbf{r}_2 \dots d\mathbf{r}_n$ is the probability of *simultaneously* finding a particle centered in volume element around $d\mathbf{r}_1$, another particle centered in volume element around $d\mathbf{r}_2$, and so on, where $\mathbf{r}^n$ represents the set $\{\mathbf{r}_1 \dots \mathbf{r}_n\}$. For statistically homogeneous media, the probability density $\rho_n(\mathbf{r}^n)$ only depends upon

- the relative distances $\mathbf{r}_{12}, \dots, \mathbf{r}_{1n}$, where $\mathbf{r}_{1i} = \mathbf{r}_i - \mathbf{r}_1$. Thus, for example, $\rho_2(\mathbf{r}_1, \mathbf{r}_2) = \rho_2(\mathbf{r}_{12})$. Note also that $\rho_1(\mathbf{r}_1) = \rho_p$, where ρ_p is the density of the particles
2. An important probability distribution function is $S_n(\mathbf{x}^n)$, defined as the probability of *simultaneously* finding n points at positions $\mathbf{x}_1, \mathbf{x}_2, \dots, \mathbf{x}_n$ in one of the phases (pore or particle). Similarly, $S_n^{(i)}(\mathbf{x}^n)$ is the probability that the n points at $\mathbf{x}_1, \mathbf{x}_2, \dots, \mathbf{x}_n$ are in phase i (pore or particle). If the porous medium is statistically homogeneous, then $S_1^{(i)}$ is simply the volume fraction ϕ_i of phase i . Moreover, $S_2(\mathbf{r}_1, \mathbf{r}_2) = S_2(\mathbf{r}_{12})$. For statistically homogeneous (or strictly stationary) porous media, the joint probability distributions are translationally invariant so that $S_n^{(i)}(\mathbf{x}^n) = S_n^{(i)}(\mathbf{x}^n + \mathbf{y})$. If a porous medium possesses phase-inversion symmetry (between the pore and particle phases), then $S_n^{(1)}(\mathbf{x}^n; \phi_1, \phi_2) = S_n^{(2)}(\mathbf{x}^n; \phi_2, \phi_1)$.
 3. The point/ q -particle distribution functions $G_n^{(i)}(\mathbf{x}_1; \mathbf{r}^q)$ characterize the probability of finding a point at $\mathbf{x}_1$ in phase i (pore or particle) and a configuration of q spheres with their centers at $\mathbf{r}^n$, respectively, with $n = q + 1$.
 4. We may also define surface–surface, surface–matrix, and surface–void correlation functions $F_{ss}(\mathbf{x}_1, \mathbf{x}_2)$, $F_{sm}(\mathbf{x}_1, \mathbf{x}_2)$, and $F_{sv}(\mathbf{x}_1, \mathbf{x}_2)$, associated with finding a point on the interface between the two phases (i.e., on the pores' walls) and another point in the pore phase, or on the interface, respectively. Other valuable information may be gained from a surface–particle correlation function F_{sp} that yields the correlations associated with finding a point on the particle–pore interface and the center of a sphere in some volume element. The functions F_{ss} and F_{sv} can be obtained from any plane cut through an isotropic medium. For homogeneous and isotropic media, such functions only depend on $|\mathbf{r}| = |\mathbf{x}_2 - \mathbf{x}_1|$. For homogeneous porous media, as $|\mathbf{r}| \rightarrow \infty$, one has $F_{sv}(\mathbf{r}) \rightarrow \Xi \phi_1$, and $F_{ss}(\mathbf{r}) \rightarrow \Xi^2$ where Ξ is the specific surface area (surface area per unit volume), and $\phi_1 = \phi$ the porosity.
 5. Two important properties of disordered porous media are the *exclusion* probabilities $E_V(r)$ and $E_P(r)$. The former is the probability of finding a region Ω_V – taken to be a d -dimensional spherical cavity of radius r centered at some arbitrary point – to be empty of any particle centers. Physically, $E_V(r)$ is the expected fraction of space available to a test sphere of radius $r - R$ inserted into the system. $E_P(r)$, on the other hand, is the probability of finding a region Ω_P – taken to be a d -dimensional spherical cavity of radius r centered at some arbitrary particle center – to be empty of other particle centers.
 6. The *void nearest-neighbor* probability density functions $H_V(r)$ and $H_P(r)$ are also useful quantities. $H_V(r)dr$ is the probability that, at an arbitrary point in the system, the center of the nearest particle lies at a distance in the interval $(r, r + dr)$, whereas $H_P(r)dr$ is the probability that, at an arbitrary particle center in the system, the center of the nearest particle lies at a distance in the interval $(r, r + dr)$.

The definitions of the void nearest-neighbor and exclusion probabilities imply that

$$E_i(r) = 1 - \int_0^r H_i(x) dx, \quad i = V, P \quad (7.4)$$

or, equivalently, $H_i(r) = -\partial E_i / \partial r$. A mean nearest-neighbor distance $\langle \ell_P \rangle$ is also defined by

$$\langle \ell_P \rangle = \int_0^\infty r H(r) dr = 2R + \int_0^\infty E_P(r) dr, \quad (7.5)$$

which is an important characteristic of a packing of spheres. Utilizing $\langle \ell_P \rangle$, one can precisely define the random-close packing fraction ϕ_{cp} in the fully-impenetrable spheres model. Specifically, ϕ_{cp} is defined as the maximum of packing fraction ϕ_2 over all the homogeneous and isotropic ensembles of the packings at which $\langle \ell_P \rangle = 2R$, where R is the spheres' radius. If the spheres are polydispersed with a mean radius $\langle R \rangle$, then one may define ϕ_{cp} in a similar manner by $\langle \ell_P \rangle = 2\langle R \rangle$. From a practical view point, ϕ_{cp} is the packing fraction at which randomly-arranged hard (impenetrable) spheres cannot be compressed any more if a hydrostatic pressure is applied to the packing. At ϕ_{cp} , the exclusion and the void nearest-neighbor probabilities are both zero for $r > 2R$. The numerical estimates for ϕ_{cp} , as mentioned earlier, are $\phi_{cp} \simeq 0.82$ and 0.64 for $d = 2$ and 3 , respectively. Note that ϕ_{cp} is different from the *closest packing fraction* that are $\pi/(2\sqrt{3}) \simeq 0.907$ and $\pi/(3\sqrt{2}) \simeq 0.74$ for $d = 2$ and 3 , respectively.

7. Two other important characteristics are the *lineal-path function* $L_p^{(i)}(z)$ (Lu and Torquato, 1992a,b) and the *chord-length distribution function* $L_c^{(i)}(z)$ (Torquato and Lu, 1993). $L_p^{(i)}(z)$ is the probability that a line segment of length z is entirely in phase i (pore or particle) when randomly thrown into the sample. For 3D media, $L_p^{(i)}(z)$ is equivalent to the area fraction of phase i (pore or particle), measured from the projected image onto a plane, a highly important problem in stereology (Underwood, 1970). Moreover, $L_p^{(i)}(0) = \phi_i$ and $L_p^{(i)}(\infty) = 0$. $L_p^{(i)}(z)$ contains information on the microscopic connectivity of a porous medium and, thus, it is a useful quantity. In stochastic geometry, the quantity $\phi_i[1 - L_p^{(i)}(z)]$ is sometimes referred to as the *linear contact distribution function* (Stoyan *et al.*, 1995).

$L_c^{(i)}(z)dz$, on the other hand, is the probability of finding a chord of length between z and $z + dz$ in phase i (pore or particle). Thus, $L_c^{(i)}(z)$ is actually a probability density. Chords represents distributions of lengths between intersections of lines with the interface between the phases, that is, with the pores' walls. Chord-length distributions are relevant to transport processes in heterogeneous porous media, for example, diffusion, radiative heat transfer (Ho and Strieder, 1979; Tassopoulos and Rosner, 1992), and flow and conduction (Krohn

- and Thompson, 1986). The quantities $L_p^{(i)}(z)$ and $L_c^{(i)}(z)$ are, in fact, closely related (see below).
8. A very general distribution function for characterizing the microstructure of a heterogeneous medium of the type we study here is $G_n(\mathbf{x}^p; \mathbf{r}^q)$, which is the n -point distribution function that characterizes the correlations associated with finding p particles centered at positions $\mathbf{x}^p = \{\mathbf{x}_1, \dots, \mathbf{x}_p\}$ and q particles centered at positions $\mathbf{r}^q = \{\mathbf{r}_1, \dots, \mathbf{r}_q\}$, with $n = p + q$. Clearly, for $q = 0$, we have $G_n(\mathbf{x}^n; \emptyset) = S_n(\mathbf{x}_n)$, where $\emptyset$ denotes the null set, and in the limit $p = 0$, $G_n(\emptyset; \mathbf{r}^q) = \rho_n(\mathbf{r}^q)$. Moreover, if $p = 1$ and the radius of the p -particles is taken to be zero, then $G_n(\mathbf{x}_1; \mathbf{r}^q) = G^{(1)}(\mathbf{x}_1; \mathbf{r}^q)$, as defined in item 3.
 9. A most general n -point distribution function $H_n(\mathbf{x}^m; \mathbf{x}^{p-m}; \mathbf{r}^q)$ is defined (Torquato, 1986) to be the correlation function associated with finding m points with positions $\mathbf{x}^m$ on certain surfaces within the medium, $p - m$ with positions $\mathbf{x}^{p-m}$ in certain spaces *exterior* to the spheres, and q sphere centers with positions $\mathbf{r}^q$ with $n = p + q$, in a statistically *heterogeneous* porous medium of N identical d -dimensional spheres. Most of the n -point correlation and distribution functions that were described above represent some limiting cases of $H_n(\mathbf{x}^m; \mathbf{x}^{p-m}; \mathbf{r}^q)$. By way of its construction, the function H_n , unlike the less general function G_n , also contain interfacial information about the available surfaces associated with the first m test particles. Note that in the cherry-pit model, $H_n(\mathbf{x}^m; \mathbf{x}^{p-m}; \mathbf{r}^q)$ is identically zero for certain $\mathbf{r}^q$. In particular, for any value of the impenetrability λ (see above), we have $H_n(\mathbf{x}^m; \mathbf{x}^{p-m}; \mathbf{r}^q) = 0$ if $|\mathbf{r}_i - \mathbf{r}_j| < 2\lambda R$, where R is the radius of the spheres.

The functions defined above are not all independent and are, in fact, related to one another. Suppose that the d -dimensional spheres are spatially distributed according to a specific N -particle probability density $P_N(\mathbf{r}^N)$. Then, $\rho_n(\mathbf{r}^n)$, which is sometimes called the n -particle *generic* probability density, is given by

$$\rho_n(\mathbf{r}^n) = \frac{N!}{(N-n)!} \int P_N(\mathbf{r}^N) d\mathbf{r}_{n+1} \cdots d\mathbf{r}_N. \quad (7.6)$$

Many of correlation and distribution functions defined above are related to the n -point distribution function H_n :

$$S_n(\mathbf{x}^n) = \lim_{a_i \rightarrow R} H_n(\emptyset; \mathbf{x}^n; \emptyset), \quad \forall i, \quad (7.7)$$

$$G_n(\mathbf{x}_1; \mathbf{r}^q) = \lim_{a_1 \rightarrow R} H_n(\emptyset; \mathbf{x}_1; \mathbf{r}^q), \quad (7.8)$$

$$F_{ss}(\mathbf{x}_1; \mathbf{x}_2) = \lim_{a_i \rightarrow R} H_2(\mathbf{x}_1, \mathbf{x}_2; \emptyset, \emptyset), \quad \forall i, \quad (7.9)$$

$$H_V(r) = H_1(\mathbf{x}_1; \emptyset, \emptyset), \quad E_V(r) = H_1(\emptyset; \mathbf{x}_1; \emptyset). \quad (7.10)$$

One can also show that $E_p(r) = H_2(\emptyset; \mathbf{x}_1; \mathbf{r}_1)/\rho_1(\mathbf{r}_1)$, as $|\mathbf{x}_1 - \mathbf{r}_1| \rightarrow 0$, from which the relation $H_p(r) = -\partial E_p / \partial r$ is obtained.

As for the lineal-path function, we focus on the quantity $L_p(z) = L_p^{(1)}$ for a two-phase (pores and particles) heterogeneous materials of interest in this book. The

key idea is that $L_p(z)$, which is a type of exclusion probability function, yields the probability of inserting a test particle – a line of length z – into the system which is equal to the probability of finding an exclusion zone $\Omega_E(z)$ between a line of length z and a sphere of radius R . The region Ω_E is a d -dimensional spherocylinder of cylindrical length z and radius R with hemispherical caps of radius R on either end of the cylinder. Therefore, if we define the exclusion indicator function $m(\gamma; z)$ by

$$m(\gamma; z) = \begin{cases} 1, & \gamma \in \Omega_E(z) \\ 0, & \text{otherwise} \end{cases} \quad (7.11)$$

then, $L_p(z)$ is given by (Lu and Torquato, 1992a,b)

$$L_p(z) = 1 + \sum_{k=1}^{\infty} \frac{(-1)^k}{k!} \int \rho_k(\mathbf{r}^k) \prod_{j=1}^k m(\mathbf{x} - \mathbf{r}_j; z) d\mathbf{r}_j. \quad (7.12)$$

(Lu and Torquato, 1993a,b) showed that for statistically isotropic two-phase materials of *arbitrary* microgeometry, the chord-length distribution function is related to the lineal-path function $L_p(z)$ by

$$L_c(z) = \frac{\ell_C}{\phi_1} \frac{d^2 L_p(z)}{dz^2}, \quad (7.13)$$

where ℓ_C is the average of $L_c(z)$,

$$\ell_C = \int_0^{\infty} z L_c(z) dz. \quad (7.14)$$

Lu and Torquato (1991) generalized Eqs. (7.7)–(7.14) to a polydispersed medium in which the spheres' radii follow a probability density function.

7.2.3

The n -Particle Probability Density

Given the n -particle probability density ρ_n , one can compute the correlation function H_n , from which most other properties follow. The advantage of this formulation is that the function ρ_n has been studied in great detail in the context of the statistical mechanics of liquids (see, for example, Hansen and McDonald, 1986) and, therefore, the extensive results obtained in such studies can be immediately employed for modeling porous media of the type we are describing here.

In the fully-penetrable sphere model – the Swiss-cheese model that represents the limit $\lambda = 0$ of the impenetrability parameter λ – with a particle density (number of particles per unit volume) ρ_p , there is no spatial correlation between the particles. Therefore, one has the *exact* relation

$$\rho_n(\mathbf{r}^n) = \rho_p^n, \quad \forall n. \quad (7.15)$$

In this case,

$$S_n(\mathbf{r}^n) = \exp(-\rho_p V_n) , \quad (7.16)$$

where V_n is the *union volume* of the n spheres (see below). Determining $\rho_n(\mathbf{r}^n)$ for other types of the dispersion of spheres is considerably more difficult. For example, for fully-impenetrable spheres, the impenetrability condition cannot by itself uniquely determine the ensemble, and one must supply more information. One must, for example, state that the spheres are distributed in the most random fashion that, together with the impenetrability condition, determines the state of the system.

Let us define

$$\Theta(x) = \begin{cases} 1, & r > 0 \\ 0, & r < 0 \end{cases} \quad (7.17)$$

and $v_2^i(r; R, R)$, the intersection volume of two identical d -dimensional spheres of radius R with their centers separated by a distance r , which is given by

$$v_2^i(r; R, R) = 2R^2 \left[\cos^{-1} \left(\frac{r}{2R} \right) - \frac{r}{2R} \left(1 - \frac{r^2}{4R^2} \right)^{\frac{1}{2}} \right] \Theta(2R - r) , \quad d = 2 \quad (7.18)$$

$$v_2^i(r; R, R) = \frac{4\pi R^3}{3} \left(1 - \frac{3r}{4R} + \frac{r^3}{16R^3} \right) \Theta(2R - r) , \quad d = 3 . \quad (7.19)$$

Determining the intersection volume of three or more spheres is non-trivial, especially if the spheres' radii are not the same (Torquato, 2002; Sahimi, 2003a).

7.2.4

Distribution of Equal-Size Particles

Consider a system of identical particles of *arbitrary* shapes with number density ρ_p . We define a dimensionless density $\eta = \rho_p v$, where v is the volume of a particle that, for example, for a d -dimensional sphere of radius R is given by

$$v(R) = \frac{\pi^{\frac{d}{2}} R^d}{\Gamma \left(1 + \frac{d}{2} \right)} , \quad (7.20)$$

where $\Gamma(x)$ is the gamma function. For fully-impenetrable particles, the reduced density η is *exactly* the particle volume fraction ϕ_2 , that is,

$$\eta = \phi_2 = 1 - \phi_1 = 1 - \phi , \quad (7.21)$$

where ϕ is the porosity. The equality does not hold if the particles can overlap. In particular, for the penetrable-concentric shell model, one has

$$\eta(\lambda) \geq \phi_2(\lambda) , \quad (7.22)$$

where λ is the impenetrability index defined above so that the equality applies when $\lambda = 1$.

7.2.4.1 Fully-Penetrable Spheres

For the fully-penetrable spheres model which represents the limit $\lambda = 0$, the first two functions S_1 and S_2 are given by

$$S_n = \begin{cases} \phi = \phi_1 = 1 - \phi_2 = \exp(-\eta), & n = 1, \\ \exp\left[-\eta \frac{V_2(r; R)}{v}\right], & n = 2, \end{cases} \quad (7.23)$$

where V_2 is the union volume of two spheres to be defined shortly. Given that for this model, the n -particle probability density function is given by Eq. (7.15), calculation of $H_n(\mathbf{x}^m; \mathbf{x}^{p-m}; \mathbf{r}^q)$ is straightforward. In particular, if we let $m = q = 0$, then $H_n(\mathbf{x}^n)$, the probability of inserting n spheres of radii $a_1, \dots, a_n$ into a system of N spheres of radius R at positions $\mathbf{x}_1, \dots, \mathbf{x}_n$, that is, inserting the particles into the available space of the medium, is given by

$$H_n(\mathbf{x}^n) = \exp[-\rho_p V_n(\mathbf{x}^n; a_1, \dots, a_n)]. \quad (7.24)$$

Here, $V_n(\mathbf{x}^n; a_1, \dots, a_n)$ is the union volume of nd -dimensional spheres of radii $a_1, \dots, a_n$, centered at $\mathbf{x}_1, \dots, \mathbf{x}_n$, respectively. In the limit that $a_i \rightarrow R$ for $\forall i$, one recovers Eq. (7.16). Note that the union volume $V_2(r_{12}; a_1, a_2)$ of two spheres is given by $V_2(r_{12}; a_1, a_2) = v(a_1) + v(a_2) - v_2^i(r_{12}; a_1, a_2)$, and

$$\begin{aligned} V_3(r_{12}, r_{13}, r_{23}; a_1, a_2, a_3) = & v(a_1) + v(a_2) + v(a_3) - v_2^i(r_{12}; a_1, a_2) \\ & - v_2^i(r_{13}; a_1, a_3) - v_2^i(r_{23}; a_2, a_3) \\ & + v_3^i(r_{12}, r_{13}, r_{23}; a_1, a_2, a_3), \end{aligned} \quad (7.25)$$

with $r_{ij} = |\mathbf{x}_i - \mathbf{x}_j|$, and v being the volume of one sphere given by Eq. (7.20).

Other properties of the model can also be determined. The model predicts that the specific surface area Ξ is given by

$$\Xi = \lim_{a_1 \rightarrow R} H_1(\mathbf{x}_1; \emptyset, \emptyset) = \rho_p \phi_1 \frac{dv}{dR} = \frac{\eta \phi_1 d}{R} = \frac{\eta \phi d}{R}, \quad (7.26)$$

where v and ϕ are given by Eqs. (7.20) and (7.23), respectively. Equation (7.41) must be compared with the corresponding results for the impenetrable sphere model: $s = \rho dv/dR$. Since there are no correlations in the model, there is no difference between the void and particle nearest-neighbor functions, $H_p(r) = H_v(r) = H(r)$. One can then show that,

$$H(r) = \rho_p \frac{dv(r)}{dr} \exp[-\rho_p v(r)], \quad (7.27)$$

where $v(r)$ is given by Eq. (7.20). Using Eqs. (7.4) and (7.27), we then obtain

$$E(r) = \exp\left[-\eta \frac{v_d(\mathbf{x})}{v_d(1)}\right], \quad (7.28)$$

where

$$v_d(\mathbf{x}) = \frac{1}{2^d} \frac{\pi^{\frac{d}{2}} \mathbf{x}^d}{\Gamma\left(1 + \frac{d}{2}\right)}. \quad (7.29)$$

One can also derive the lineal-path function $L_p(z) = L_p^{(1)}(z)$ for this model. Using Eqs. (7.12) and (7.15), it is not difficult to see that

$$L_p(z) = \exp[-\rho_p v_E(z)] , \quad (7.30)$$

where $v_E(z)$ is the d -dimensional volume of the exclusion zone Ω_E defined by Eq. (7.11), which in d dimensions is given by

$$v_E(z) = \frac{\pi^{\frac{d}{2}} R^d}{\Gamma\left(1 + \frac{d}{2}\right)} + \frac{\pi^{\frac{d-1}{2}} R^{d-1}}{\Gamma\left[1 + \frac{d-1}{2}\right]} z . \quad (7.31)$$

Once $L_p(z)$ is known, the chord-length distribution $L_c(z)$ can be immediately computed using Eq. (7.13). Utilizing Eq. (7.23), we can rewrite the results for $L_p(z)$ in terms of the volume fraction of the pores, that is, the porosity $\phi = \phi_1 = 1 - \phi_2$, with the results being

$$L_p(x) = \begin{cases} \phi^{1+(4/\pi)x} , & d = 2 , \\ \phi^{1+(3/2)x} , & d = 3 , \end{cases} \quad (7.32)$$

where $x = z/(2R)$. Equations (7.32) indicate that with increasing z , the lineal-path function decreases with the porosity since it becomes increasingly more difficult to insert a line segment of length z in the pore space.

7.2.4.2 Fully-Impenetrable Spheres

Unlike the case of fully-penetrable spheres, the fully-impenetrable sphere model is much more difficult to analyze because there are significant correlations in the system that are imposed by the impenetrability condition. Despite this difficulty, significant progress has been made that has been described by Torquato (2002) and Sahimi (2003a).

In general, one must have $E_p(r) = 1$ and, $H_p(r) = 0$ for $0 \leq r < 2R$. It can also be shown that $E_v(r) = 1 - \rho_p v(r)$ and $H_v(r) = \rho_p s$ for $0 \leq r \leq R$, where v is the volume of a cavity of radius r , and s is the specific surface area. In general, the functions $H_p(r)$ and $H_v(r)$ are not truncated series, but are expressed as an infinite series. For $d = 2$, an accurate approximation to $H_p(x)$ is given by Torquato *et al.* (1990)

$$H_p(x) = \frac{4\eta(2r - \eta)}{(1 - \eta)^2} \exp \left\{ -\frac{4\eta}{(1 - \eta)^2} [x^2 - 1 + \eta(x - 1)] \right\} , \quad x > 1 , \quad (7.33)$$

where $\eta = \rho_p v$ is the dimensionless density (volume fraction of the particles) defined above, and $x = r/(2R)$. For $d = 3$, we have

$$H_p(x) = 24\eta (f_1 + f_2 x + f_3 x^3) \times \exp \left\{ -\eta [24f_1(x - 1) + 12f_2(x^2 - 1) + 8f_3(x^3 - 1)] \right\} , \quad x > 1 , \quad (7.34)$$

with

$$f_1 = \frac{1}{2}\eta^2 f(\eta), \quad f_2 = -\frac{1}{2}\eta(3 + \eta)f(\eta), \quad f_3 = (1 + \eta)f(\eta), \quad (7.35)$$

where $f(\eta) = (1 - \eta)^{-3}$. Given Eqs. (7.34) and (7.35) for $H_P(r)$ and $H_V(r)$, the corresponding equations for $E_P(r)$ and $E_V(r)$ can be immediately determined using Eq. (7.4). Accurate approximations for $L_p(z)$ are given by (Lu and Torquato, 1992a,b)

$$L_p(z) = (1 - \eta) \exp \left[-\frac{d\eta z}{\beta R(1 - \eta)} \right], \quad (7.36)$$

where $\beta = \pi$ and four for $d = 2$ and three, respectively. Finally, it can be shown that for any ergodic ensemble of isotropic packings of identical d -dimensional hard spheres, the mean nearest-neighbor distance $\langle \ell_P \rangle$, defined by Eq. (7.5), is bounded from above: $\langle \ell_P \rangle \leq 1 + (2^d \eta d)^{-1}$.

7.2.4.3 Interpenetrable Spheres

Using computer simulations, Lee and Torquato (1988) calculated the pore volume fraction (porosity) $\phi = \phi_1$ in the penetrable concentric shell model as a function of the parameter λ . The following approximate, but very accurate, formulae for ϕ were derived by Rikvold and Stell (1985) that are in excellent agreement with the numerical results of Lee and Torquato (1988):

$$\phi(\eta, \lambda) = (1 - \lambda^d \eta) \exp \left[-\frac{(1 - \lambda^d)\eta}{(1 - \lambda^d \eta)^d} \right] \Psi_d(\eta, \lambda), \quad (7.37)$$

where $\Psi_d(\eta, \lambda)$ is a d -dependent function given by

$$\Psi_2(\eta, \lambda) = \exp \left[-\frac{\lambda^2 \eta^2 (1 - \lambda)^2}{(1 - \lambda^2 \eta)^2} \right], \quad (7.38)$$

$$\Psi_3(\eta, \lambda) = \exp \left[-\frac{3\lambda^3 \eta^2}{2(1 - \lambda^3 \eta)^3} (2 - 3\lambda + \lambda^3 - 3\lambda^4 \eta + 6\lambda^5 \eta - 3\lambda^6 \eta) \right]. \quad (7.39)$$

Note that $\eta \lambda^d$ represents the *hard-core* volume fraction in d dimensions.

7.2.5

Distribution of Polydispersed Spheres

For porous media applications, a more realistic version of a model of dispersion of spheres is one in which the radii of the spheres are distributed according to a normalized probability density function $f(R)$. Polydispersity leads to more flexibility in the model and, hence, can be exploited for a variety of purposes.

7.2.5.1 Fully-Penetrable Spheres

For this class of models, the volume fraction of the pores, that is, the porosity, can be obtained from a modification of Eq. (7.23) (Chiew and Glandt, 1984),

$$S_1 = \phi_1 = \phi = \exp [-\rho_p \langle v(R) \rangle] , \quad (7.40)$$

and the specific surface s area is given by

$$\Xi = \rho_p \frac{\partial \langle v(R) \rangle}{\partial R} \exp [-\rho_p \langle v(R) \rangle] . \quad (7.41)$$

Stell and Rikvold (1987) showed that

$$S_n(\mathbf{x}^n) = \exp [-\rho_p \langle V_n(\mathbf{x}^n; R, \dots, R) \rangle] , \quad (7.42)$$

where V_n is the union volume of n spheres of radius R defined and discussed earlier.

The lineal-path function has also been determined for this class of models. In order to compute this function, the exclusion indicator function defined by Eq. (7.11) must be generalized for a polydispersed system. In this case, one defines an exclusion *region* indicator function by

$$m_j(\mathbf{x}; z) = \begin{cases} 1, & \mathbf{x} \in \Omega_E(z, R_j) \\ 0, & \text{otherwise,} \end{cases} \quad (7.43)$$

where R_j is the radius of the j th sphere. The lineal-path function $L_p(z) = L_p^{(i)}(z)$ is then given by (Lu and Torquato, 1993a,b),

$$L_p(z) = 1 + \sum_{k=1}^{\infty} \frac{(-1)^k}{k!} \int dR_1 \dots dR_k f(R_k) \rho_k(\mathbf{r}^k; R_1, \dots, R_k) \prod_{j=1}^k m_j(\mathbf{x}; z) d\mathbf{r}_j , \quad (7.44)$$

which is a generalization of Eq. (7.12) to polydispersed systems. Then, using Eqs. (7.15) and (7.44), one obtains

$$L_p(z) = \exp [-\rho_p \langle v_E(z, R) \rangle] , \quad (7.45)$$

where $v_E(z, R)$ is given by Eq. (7.31). Combining Eqs. (7.23), (7.31) and (7.45), and proceeding in the same manner as that for the equal-sized particles, we obtain the analogs of Eqs. (7.32) for polydispersed systems:

$$L_p(z) = \begin{cases} \phi^{1 + \frac{2\langle R \rangle z}{\pi \langle R^2 \rangle}} , & d = 2 , \\ \phi^{1 + \frac{3\langle R^2 \rangle z}{4\langle R^3 \rangle}} , & d = 3 , \end{cases} \quad (7.46)$$

where ϕ is the porosity.

7.2.5.2 Fully-Impenetrable Spheres

Fully-impenetrable sphere models have many intriguing properties, many of which are not well-understood yet (Torquato, 2002; Sahimi, 2003a). Very little is also known about the packing characteristics of polydisperse hard spheres. For example, even if we consider one of the simplest of such polydisperse packings, namely, a *binary* mixture of hard spheres of arbitrary radii R_1 and R_2 , its largest achievable packing fraction is not known. Despite such difficulties, some progress has been made which is now summarized. For polydisperse impenetrable spheres, the following results are known:

$$S_1 = \phi_1 = \phi = 1 - \rho_p \langle v(R) \rangle, \quad (7.47)$$

where ϕ is the porosity, and the specific surface area s is given by

$$\Xi = \rho_p \frac{d \langle v(R) \rangle}{dR} = d\eta \frac{\langle R^{d-1} \rangle}{\langle R^d \rangle}. \quad (7.48)$$

Accurate approximations for the lineal-path function $L_p(z)$ have also been derived for this class of models, with the result for a d -dimensional system being

$$L_p(z) = \phi \exp \left[- \frac{\Gamma(1 + \frac{1}{2}d) \eta \langle R^{d-1} \rangle}{\pi^{\frac{d}{2}} \phi \langle R^d \rangle} \right]. \quad (7.49)$$

7.2.6

Simulation of Packings of Spheres

What is the most efficient method for computer simulation of a packing of spherical particles? If the spheres are allowed to overlap, then the simulation is rather straightforward. The problem is more difficult if one wishes to model the fully-impenetrable spheres model, or to take into account the effect of the hard cores that the spheres may have (represented by the impenetrability parameter λ).

The classical method of computer generation of a packing of spherical particles in the fully-impenetrable spheres model is that of Visscher and Bolsterli (1972). In their algorithm, a sphere is dropped into the simulation box from the top. If the particle hits the “floor”, it stops. If it hits another particle, say p_1 , it rolls down on p_1 until it hits another particle p_2 . Then, it rolls on both p_1 and p_2 until it hits particle p_3 . If its contact with p_1 , p_2 , and p_3 is stable, it stops. Otherwise, it rolls on the double contacts, and so on. Since the particle’s motion is always downward, the effect of the gravity is automatically taken into account.

As a simple method that takes into account the effect of the hard core of spherical particles, consider, for example, a system of d -dimensional spheres. The number N of the particles and the volume Ω of the system are fixed. The particles are initially placed in a cubical cell (with volume $\Omega = L^d$) on the sites of a regular lattice, for example, the face-centered or body-centered lattice. No hard-core overlap is assumed initially. The particles are then moved randomly by a small distance to new positions. The new positions are either accepted or rejected according to whether

or not the hard cores overlap. One usually uses *periodic boundary conditions* that means that, if a particle exits from an external face of the system, an identical particle enters the system from the opposite face of the system. This type of boundary condition eliminates the boundary effects, hence allowing one to simulate an essentially infinitely-large packing. After the particles have been moved a sufficiently large number of times, the system reaches equilibrium and its configuration no longer changes.

7.3

Models Based on Diagenesis of Porous Media

A review of the literature indicates that there have been a few attempts to model the diagenetic processes that have given rise to the present rock, and in particular oil reservoirs. Among such attempts the work of Roberts and Schwartz (1985) is notable. They developed a geometrical model for sandstones – usually referred to as the grain-consolidation model – that mimics many features of the natural ones. In their model the initial porous medium (before the diagenetic processes began) is a dense pack of randomly-distributed spherical grains of radius R . The model was originally suggested by Bernal (1959, 1960); see also Alben *et al.* (1976) for studying of liquids. In the model of Roberts and Schwartz (1985) the coordinates of the spheres' centers followed the Bernal distribution. Figure 7.4 shows the initial dense packing of the grain with the volume fraction of the spheres being 0.636 and, therefore, an initial porosity of 0.364.

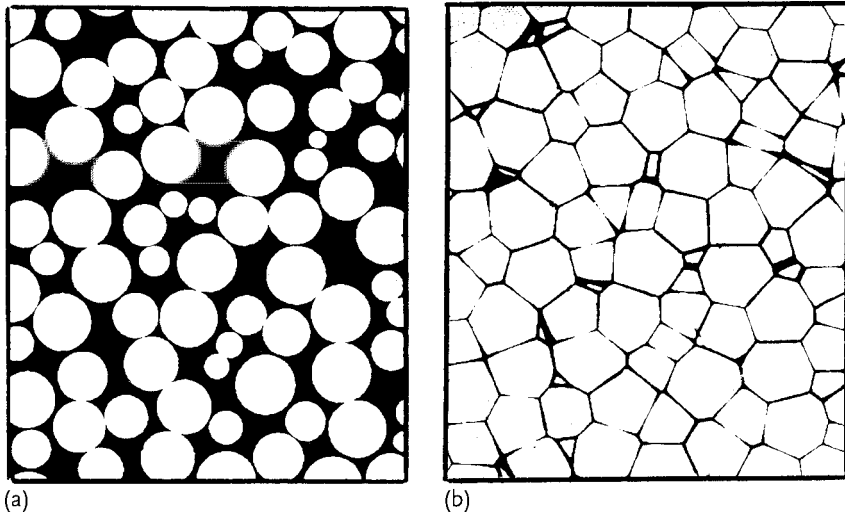


Figure 7.4 Two stages of the grain-consolidation model with porosities $\phi = 0.36$ (a) and $\phi = 0.03$ (b). Dark areas represent the pores (after Schwartz and Kimminau, 1987).

The radii of the spheres are then allowed to increase *simultaneously*, as a result of which the porous medium's porosity and effective permeability both decrease. The growth of the particles is continued until a desired porosity is reached. The percolation threshold of the porous medium, that is, the critical porosity below which no fluid can flow macroscopically in the porous medium, was determined to be $\phi_c \simeq 0.03 \pm 0.004$. The porous medium at the critical porosity is also shown in Figure 7.4. Such a low critical porosity implies that the grain-consolidation model can generate models of porous media in which the porosity can vary over more than one order of magnitude. The model shown in Figure 7.4 also has striking resemblance to natural sandstone, shown in Figure 7.5. Given that the porosity of sandstone is typically less than 0.4, the grain-consolidation model generates reasonably accurate representation of such natural porous media. The model was further studied by Schwartz and Kimminau (1987) and by Schwartz *et al.* (1989). In particular, the latter group utilized an initial dense packing of nonspherical grains in order to generate a model of anisotropic or stratified porous media.

An advantage of the grain-consolidation model is its flexibility. If, instead of the Bernal distribution, one begins with a simple-cubic lattice of spherical grains and utilizes the algorithm for the growth of the particles, then the critical porosity of the medium will be 0.349, which is close to that of the random sphere packing. On the other hand, if one begins with a body-centered cubic lattice of spherical grains, the critical porosity will be 0.0055, one order of magnitude smaller than what can be achieved with the Bernal distribution.

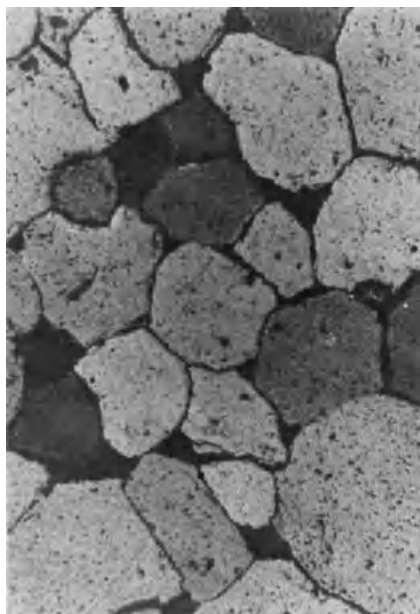


Figure 7.5 Cemented Devonian sandstone from Illinois. Compare this with Figure 7.4b (courtesy of Dr. L.M. Schwartz).

Moreover, the sedimentation and diagenetic processes that give rise to many sedimentary rocks, such as sandstones, tend to favor a distribution of particles that are roughly equal in size (Pittmann, 1984). Thus, from this perspective, the grain-consolidation model is much more efficient than a model in which the porosity is reduced by adding additional spherical particles with smaller and smaller sizes, in order to progressively fill the pore space of the original packing. In fact, to obtain a comparable porosity range by adding smaller particles, one must use spheres with radii that vary over many orders of magnitude. Even then, the final result has very little resemblance to most naturally occurring granular porous media.

While the grain-consolidation model is intended for sandstones, similar and perhaps more advanced models for other types of rock are also needed. Biswal *et al.* (2007) developed a multiscale model for the diagenesis of carbonate rock. Carbonate porous media are characterized by a wide pore size distribution, 2–3 orders of magnitude variations in their effective permeability (at the same porosity) and, perhaps most importantly, the dependence of their properties on the resolution. In addition, their morphology contains extended correlations. The model by Biswal *et al.* (2007) captures all the three characteristics. It uses 2D sections of the sample porous medium, and is based on assuming that the rock is random, but with a correlated sequence of points that are decorated with crystallites which are convex sets, for example, spheres or polyhedra. All the properties can be computed at arbitrary resolution, which offers a grain advantage over other models.

7.4

Reconstruction of Porous Media

An emerging method for developing a model of a porous medium is by *reconstruction*: Given a set of data for some properties of a porous medium, one tries to develop a model that reproduces the data most accurately by somehow minimizing the differences between the calculated and target data. The idea was first developed by Joshi (1974) and later by Quiblier (1984). It consisted of determining the first two moments – the porosity and two-point correlation function (see Section 4.17.3) – from 2D thin section images of the sample. Then, stochastic 3D samples are generated such that they match the measured statistical properties of the sample. Adler *et al.* (1990, 1992) used such a technique to reconstruct Fontainebleau sandstone. The computed effective permeability and electrical conductivity were in fair agreement with the data, but also consistently smaller than them. As Øren and Bakke (2002) pointed out, the discrepancy was due to the fact that the percolation threshold of the sandstone is around 0.02–0.03, whereas the reconstructed stochastic models had a percolation threshold of around 0.1. Higher-order statistics were used by Okabe and Blunt (2005) in reconstruction of porous media. Keehm *et al.* (2004) used data from thin sections to carry out a 3D reconstruction. They then used the lattice Boltzmann approach (see Chapters 9–12) to compute the effective permeability, which was found to be in agreement with the measured data.

One method of reconstruction is based on a stochastic optimization method by which the sum of the squared differences between the computed and measured data is minimized. Several optimization methods are currently available (Sahimi and Hamzehpour, 2010). Notable among them are the gradient-based optimizers (GBOs), the genetic algorithm (GA), and the simulated-annealing (SA) method. The GBOs are deterministic methods, whereas the SA and GA are both stochastic techniques. Among other disadvantages of the GBOs (see, for example, Deschamps *et al.*, 1998) are the fact that they might get trapped in the local minima of the energy function, and might also produce incorrect results for complex energy functions of the type given by Eq. (7.51) below. They also require computation of the derivatives of the functions, thus making them expensive as well as accurate initial guesses for the parameters to be optimized because, otherwise, the optimization technique might produce different results for different initial guesses.

Thus, the SA method (Kirkpatrick *et al.*, 1983) and the GA (Reeves and Rowe, 2003) have been utilized frequently in the reconstruction methods. Let us define an “energy” E by

$$E = \sum_{j=1}^n [f(\mathbf{O}_j) - f(\mathbf{S}_j)]^2, \quad (7.50)$$

where $\mathbf{O}_j$ and $\mathbf{S}_j$ represent the observed (or measured) and the corresponding simulated (or calculated) properties of the porous medium, respectively, with n being the number of data points. If there are more than one set of data for *distinct* properties of the porous medium, the energy E is generalized to

$$E = \sum_{i=1}^m W_i E_i, \quad (7.51)$$

where E_i is the total energy for the data set i , defined by Eq. (7.50), and W_i is the corresponding weight, as two distinct set of data for the same porous medium do not have the same weight or significance.

To initiate the reconstruction process, one begins with an initial guess for the structure of the porous medium and uses the SA method or the GA or any other suitable technique. For example, in the SA approach, two randomly-selected points of the model are then interchanged. For example, if the model is in the form of a pore network, two pores are selected at random and their radii are interchanged. The new energy E' of the system and the energy difference, $\Delta E = E' - E$, are then computed. The interchange is then accepted with a probability $p(\Delta E)$. Then, according to the Metropolis algorithm,

$$p(\Delta E) = \begin{cases} 1, & \Delta E \leq 0, \\ \exp\left(-\frac{\Delta E}{T}\right), & \Delta E > 0, \end{cases} \quad (7.52)$$

where T is a fictitious temperature.

Statistical mechanics of thermal systems provides the theoretical foundation for such an algorithm. We know that if a system is heated up to a high temperature T

and then slowly cooled down to absolute zero, the equilibrium state of the system will be its ground state. The cooling, usually called the *annealing schedule*, is selected to be sufficiently slow so as to allow the system to reach its true equilibrium state rather than getting trapped in a local energy minimum (a metastable state), and attain its true global minimum energy. At each annealing step i , the system is allowed to evolve long enough to “thermalize” at $T(i)$. We then lower T according to a prescribed annealing schedule. The cooling continues until the true ground state of the system is reached within some acceptable error, that is, when E is deemed to be small enough. This concludes the reconstruction process.

Such a procedure was utilized by Yeong and Torquato (1998a,b), Cule and Torquato (1999), Manwart and Hilfer (1999), and Sheehan and Torquato (2001) for reconstructing various models of random media. Manwart *et al.* (2000) used the method to reconstruct sandstones, given some data for the porous medium. Talukdar and Torsaeter (2002) and Talukdar *et al.* (2002a,b) used the SA method to reconstruct a variety of porous media, including chalk. Levitz (1998) reviewed methods of off-lattice reconstruction of porous media.

Another reconstruction method is based on mimicking the actual processes that have given rise to the present natural porous media, and in particular, sedimentary rock. We already described one such reconstruction method, namely, the grain-consolidation model. Bakke and Øren (1997) developed a more advanced version of the method by incorporating grain size distribution and other petrographical data obtained from 2D thin sections. Their reconstruction model attempts to mimic three fundamental processes that contribute to the formation of sandstones, namely, sedimentation, compaction, and diagenesis. Øren *et al.* (1998) applied the method to reconstructing Bentheimer sandstone, Øren and Bakke (2002) did the same for Fontainebleau sandstone, while Øren and Bakke (2003) reconstructed the Bera sandstone. In all cases, they successfully predicted the flow and transport properties, including the drainage and imbibition relative permeabilities (see Chapter 15) to two-phase flow in the porous medium.

To model sedimentation, the measured grain size distribution was used. Grains were picked at random from their size distribution and dropped into a predefined box using a sequential deposition algorithm that simulated the successive deposition of the individual grains in a gravitational field. Each grain falls under its own weight and, after hitting the surface, tries to find the most stable position to settle in.

The compaction process, which plays an important role in decreasing the porosity, was modeled as a linear process in which the vertical ordinate of every sand grain was shifted downwards according to

$$z = z_0 (1 - \beta_c) , \quad (7.53)$$

where z_0 is the initial position. Here, $0 \leq \beta_c \leq 1$ is a compaction parameter that controls the amount of compaction.

A diagenetic process was modeled based on the work of Schwartz and co-workers who developed the grain consolidation model described earlier. Thus, quartz ce-

ment growth of particles with initial radius R_0 was modeled according to

$$R(\mathbf{r}) = R_0 + \min[al(\mathbf{r})^\gamma, l(\mathbf{r})]. \quad (7.54)$$

Here, $l(\mathbf{r})$ is the distance between the surface of the original spherical grains and the surface of its Voronoi polyhedron (see Chapter 3 for the relation between the packing of spherical particles and the Voronoi polyhedra) along the direction $\mathbf{r}$. The constant a controls the amount of cement growth and, hence, the porosity, and γ controls the direction of growth. For $\gamma > 0$, the growth of the quartz cement in the direction of large $l(\mathbf{r})$ is favored (i.e., pore bodies shrink), whereas the opposite is true for $\gamma < 0$, that is, in the direction of the pore throats, which means that they are blocked off with the cement growth. The growth increases the tortuosity and leads to nonzero percolation threshold for the porosity, which was already mentioned when we described the grain consolidation model.

Two other ingredients were included in the reconstruction method. One was the local porosity distribution, first suggested by Hilfer (1991a, 1992); see also Hilfer (1996) and Biswal *et al.* (1998, 1999), and described briefly in Chapter 4. The essential idea is to measure geometric observables within a finite subset of a porous medium and then to prepare their histograms or distributions. Then, the local porosity distribution represents the probability of finding the local porosity in the interval $(\phi - \Delta\phi, \phi + \Delta\phi)$, which is similar to spatial distribution of the porosity (and other characteristics) of the FS porous media described in Chapter 5, except that the size of the cells within which the porosity is measured could be quite small and, therefore, applicable to even small-scale samples.

To incorporate the effect of the connectivity in their reconstruction, Øren and Bakke used the concept of local percolation probabilities introduced by Hilfer (1991a, 1996). The local percolation probability P_i is the fraction of measurement cells of sidelength L and local porosity ϕ that percolates in the direction i .

Hilfer and Manwart (2001) computed the effective permeability and conductivity of several reconstructed models of a Fontainebleau sandstone, and compared the results with the data. They reported that although the geometrical properties of the models were similar, the flow and transport properties were not. Only the reconstruction model of Øren and Bakke described earlier provided accurate estimates of the flow and transport properties. Hilfer and Manwart (2001) stated that the reason for the discrepancies between the various models is apparently due to the truncation of the correlations in the reconstruction process. A more detailed analysis of Fontainebleau sandstone and comparison with reconstruction models was made by Manwart *et al.* (2002) for several models. The conclusion was that the reconstruction model that takes into account the diagenesis of porous media also produce accurate predictions for their flow and transport properties. Most recently, Latief *et al.* (2010) developed a model of Fontainebleau sandstone using 2D images, which represents an extension of the model of Biswal *et al.* (2007) briefly described earlier. See also Thovert *et al.* (2001) for an earlier reconstruction model.

Another comparison between reconstruction models of Berea sandstone and the actual data was made by Biswal and Hilfer (1999). They computed the local porosity distributions of the model and compared the results with the data.

7.5

Models of Field-Scale Porous Media

At the next level of complexity are field-scale (FS) porous media. The discussion in Chapter 4 should have made it clear that small- or laboratory-scale porous media can be analyzed in great detail, and reasonable understanding of their structure and properties can be obtained. This is not, however, the case with the FS porous media. As described in Chapter 6, experimental data for the important properties of such porous media are usually grossly incomplete, their flow properties, such as the effective permeability, vary greatly over distinct length scales by at least a few orders of magnitude, and the relations between such spatially-varying properties and the volume or the relevant length scales are often unknown. Such complexities make the transition from models of small porous media to the FS type very difficult.

Even if detailed information about the variability of all the quantities of interest at the FS is available, we still may not be able to obtain a reasonably complete understanding, and meaningful average properties of the porous medium at large length scales. Due to such reasons, modeling of the FS porous media usually involves stochastic techniques. The reason for using stochastic techniques is that although the FS porous media are, in principle, intrinsically deterministic due to the reasons just discussed, one often must think of them in stochastic terms and describe their properties in terms of statistical quantities because there are never enough data to reduce the uncertainty to negligible levels. By the same token, a continuum approach to fluid flow and transport in FS porous media based on the classical equations of flow and transport is often unsuccessful because such equations can provide information only about quantities that vary in a deterministic manner. One can, of course, modify the continuum approach by developing *stochastic equations* of flow and transport in which one or more variables, such as the permeability, porosity, and the fluid velocity vary stochastically. We will describe such approaches in Chapter 11, where we study transport and dispersion in porous media. Here, it suffices to point out that two major problems restrict the usefulness of stochastic equations of flow and transport. One is that the appropriate form of the governing stochastic equations is not often obvious. The second problem is that even if one knows and uses the appropriate stochastic equations, taking into account the effect of long-range correlations that often exist in the FS porous media (see Chapter 5) is very difficult.

Due to such difficulties, stochastic discrete models have been developed for describing the FS porous media. Such models represent, in some sense, an extension of the network models for small-scale porous media, except that in such models, a block of the network represents a portion of the porous medium at a length scale over which information is available. Moreover, it is relatively straightforward to include the effect of long-range correlations in such discrete models. A comprehensive discussion of such models would be too long to be given here. The interested reader should consult Haldorsen *et al.* (1988). Here, we briefly describe four basic approaches to modeling of FS porous media. Most of such approaches model 2D porous media, as the thickness of the FS porous media in which fluid flow and

transport take place is usually small compared with their length or width and, thus, such porous media are essentially 2D system. It is, however, not difficult to extend such approaches to 3D FS porous media.

7.5.1

Random Hydraulic Conductivity Models

In this approach, the 2D porous medium is represented by a rectangle which is discretized into many smaller rectangular blocks that are supposed to represent a portion of the medium that is homogeneous on the scale of the block's size. To each block, a randomly-selected hydraulic conductivity is assigned. This type of model was pioneered by Warren and Skiba (1964) and Heller (1972). In both studies, it was assumed that there are no correlations between the conductivities of the blocks. Schwartz (1977) modified the model by inserting blocks of lower conductivities in an otherwise homogeneous 2D region. One can also accommodate a non-random spatial structure by controlling the density and mode of aggregation of the inserted blocks. In principle, the blocks do not have to be rectangular.

Smith and Freeze (1979) and Smith and Schwartz (1980, 1981a,b) modified this basic model by including correlations between the blocks' hydraulic conductivities that usually exist in the FS porous media (see Chapter 5). In their model, it was assumed that the spatial variations of the hydraulic conductivities are described by a statistically-homogeneous stochastic process. The spatial structure of the conductivity field was represented by a first-order nearest-neighbor stochastic process. It was assumed that the hydraulic conductivity g_b of the blocks is log-normally distributed but, of course, any other distribution may also be used. If $Y = \log g_b$, then the first-order nearest-neighbor stochastic process implies that Y_{ij} , the random variable for the block whose center's coordinates are (i, j) , is given by

$$Y_{ij} = \alpha_x(Y_{i-1,j} + Y_{i+1,j}) + \alpha_z(Y_{i,j-1} + Y_{i,j+1}) + \epsilon_{ij}, \quad (7.55)$$

where α_x and α_z are, respectively, autoregressive parameters that express the degree of spatial dependence of Y_{ij} on its two neighboring values in the x and z directions, and ϵ_{ij} is a normal random variable uncorrelated with all the other ϵ_{ij} . If $\alpha_x = \alpha_z$, then the medium has a statistically-isotropic covariance structure. Otherwise, the porous medium is anisotropic and the covariance between the conductivity values is dependent upon the orientation. The random variables ϵ_{ij} are distributed according to a normal distribution with a zero mean and a given variance.

7.5.2

Fractal Models

The models developed by Smith and Freeze (1979) and Smith and Schwartz (1980, 1981a,b) that contained short-range correlations between the hydraulic conductivities of the blocks were significantly generalized by many others in which the permeability and porosity of the blocks followed fractal statistics and, therefore, contained

correlations with an extent that is comparable with the linear size of the system (see Chapter 5). As described in Chapter 5, this type of model was first suggested by Hewett (1986) who showed that the vertical porosity logs may be represented accurately by a fractional Gaussian noise (FGN), while the lateral distribution of the porosity may follow the statistics of a fractional Brownian motion (FBM). The two stochastic distributions induce long-range correlations in the property values, characterized by a Hurst exponents $0 < H < 1$ (see Chapter 5). A FBM with $H > 0.5$ induces *positive* or *persistent* correlations, while one with $H < 0.5$ generates *negative* or *anti-persistent* correlations. The limit $H = 1$ represents a totally smooth distribution.

7.5.3

Multifractal Models

Multifractal models were suggested by Meakin (1987) and Lenormand *et al.* (1990) as a tool for generating highly heterogeneous porous media with long-range correlations. Consider a 2D system, for example, a square grid, and a probability p that can be related at the end of the construction of the model to a measure, such as, the permeability or porosity, and is distributed uniformly in the interval $(1 - a, 1 + a)$ with $0 \leq a \leq 1$. In the first step of constructing the model, a value p_{11} is selected at random and is attributed to all the pixels of the initial square. The first dichotomy is then carried out to make four squares of size $2^{n-1} \times 2^{n-1}$ and four values, p_{21} , p_{22} , p_{23} , and p_{24} , are selected at random and attributed to each of the four squares. The same procedure is repeated n times. At the end of the process, each pixel is characterized by n values of the probability p . A new measure P is then defined that is the product of the n random values of p .

The main property of this process is that similar to any fractal distribution, it introduces correlations between pixels at all scales. A lower cut off can also be introduced into the model by considering m steps of dichotomy, where $m < n$ and n is the dimension of the pattern. One first makes $2^{n-m} \times 2^{n-m}$ independent multifractal patterns of size m_0 , and then computes $2^{n-m} \times 2^{n-m}$ independent products $p_{m+1} \times p_{m+2} \times \dots \times p_n$. The pixel values of each multifractal are then multiplied by the products in order to obtain products of order n . This procedure can be further generalized to anisotropic media by considering two probabilities p_x and p_y for the x - and y -axes, distributing p_x and p_y uniformly in $(1 - a_x, 1 + a_x)$ and $(1 - a_y, 1 + a_y)$, and identifying p by the product $p_x \times p_y$.

For simulating flow in a FS porous medium, the measure p may be thought of as the effective permeability of a portion of the porous medium. Given a pattern and a permeability distribution, flow and displacement processes in such a porous medium can be simulated and investigated. Mukhopadhyay and Sahimi (2000) used the multifractal model to study flow in the FS porous media.

7.5.4

Reconstruction Methods

Just as one may develop a model of a small-scale porous medium by a reconstruction method that was described in Section 7.4, one may utilize the same for developing a model for a FS porous medium. The difference is that a FS porous medium may require a much larger grid and, therefore, determining the global minimum of the energy function defined by Eq. (7.50) or (7.51) is a much more difficult task.

The simulated annealing (SA) method has been used in the past for reconstructing models of the FS porous media, for example, oil reservoirs. For example, Panda and Lake (1993) used the SA method together with a parallel computational strategy in order to reconstruct a model of an oil reservoir by optimizing its permeability field. Quenes and Saad (1993) also used the SA method for oil reservoir characterization, utilizing a parallel computational strategy, as did Savioli *et al.* (1996) and Saccomano *et al.* (2001). Sen *et al.* (1995) compared the performance of the SA and GA methods for stochastic reservoir modeling. Both methods were able to produce major features of the reservoir's permeability distribution, and generated the appropriate pressure profiles.

The main disadvantage of the SA method is that it cannot be used easily in massively-parallel computations as efficiently as the GA can be. Although algorithms have been developed for this purpose (see, for example, Bevilacqua, 2002; Ye and Lin, 2006), they have yet to be tested for the type of large and complex optimization problems that one must tackle for reconstructing a FS porous medium.

The GA (see, for example, Reeves and Rowe, 2003) has also been utilized in the reconstruction of the FS porous media. For example, Romero *et al.* (2000) and Romero and Carter (2001) used a GA to develop a model of an oil reservoir by optimizing the performance of a flow simulator that computed the pressures and water flow rates at a number of extraction wells in the reservoir. Sanchez *et al.* (2007) utilized the GA to reconstruct a model of a landfill, which is a large-scale porous medium with the additional complexity that, in addition to flow and transport, biodegradation and reactions with nonlinear kinetics also occur there.

One important advantage of the GA over other optimization techniques is that it is not sensitive to the initial guesses that one utilizes for the parameters that are to be determined and optimized, for example, the spatial distributions of the permeabilities and porosities. Thus, even an arbitrary initial guess may be employed by the GA. The GA ensures that practically the entire search domain is tested for optimality and, therefore, the probability of finding the global minimum of the objective function can, in principle, approach one.

Another advantage that the GAs have over many deterministic optimization techniques is that there is no need for calculating any derivative of the energy function, which can be computationally difficult and expensive if the function is complex. One disadvantage of the GAs is that its use can be computationally expensive if computing the energy function is time consuming, so much so that some optimization problems might require many weeks or even months for completion. However, the GAs spend up to 99% of the total computation time to evaluate the energy

function. Due to this fact, utilizing high-performance computers and massively-parallel computational strategies alleviates the problem.

Since the reconstruction method based on the SA algorithm was already described in Section 7.4, it may be useful to also describe the GA. The description and discussion of the GA that we present below follow those given by Sanchez *et al.* (2007).

7.5.4.1 The Genetic Algorithm for Reconstruction

Any optimization problem that utilizes a GA involves four steps:

1. the *selection* process for generating solutions;
2. the *design* of the “genome” to constrain the variables that define a possible solution, and the generation of the “phenotype” which, in the present problem, is the model of transport and reaction that we use;
3. the *crossover* and *mutation* operations that are used for generating new solutions and approaching the true optimal one, and
4. *eliticism*, which selects those solutions that eventually lead to the global minimum of the energy function.

Each time we carry out all four sets of operations, we have completed one *generation* of the computations. Let us briefly describe the various steps that are involved in using the GAs.

Selection: In Darwin’s theory, species that can adapt to their environment are selected to produce the next generation of the offsprings. In reconstruction of a FS porous medium, each specie is the set of all the parameters that we identify for determining their optimal values, given some data for one or more properties of the medium. Such parameters usually include the spatial distributions of the permeabilities and porosities. Selection is the method by which the species are chosen to produce the offsprings – the updated species for the next generation. Except for the first generation (the first trial species), selection of the species in the GAs is based on the evaluation of the energy function defined by Eq. (7.50) or (7.51). The species with a smaller energy function possesses a greater probability of producing one or more offsprings for the next generation. The set of all the possible species that we examine during the optimization process is called the *population*. One must specify the population’s size at the beginning of the optimization process.

As mentioned above, the GAs are not sensitive to the initial guesses (the initial population of the species) for the parameters to be optimized. Therefore, the first population of the species may be generated randomly in the ranges over which the parameters to be optimized are expected to vary. Of course, if one has some additional insights into the spatial distributions of the parameters to be optimized, they are also used for generating more accurate initial species and their population.

Using the population of the species, we use a model for numerical simulation of the phenomenon in the porous medium for which there are some data. For example, if the data are the time-dependent pressures at one or more production

wells in an oil reservoir, we solve the equation for the pressure transient analysis in order to compute the time-dependent pressures at the production wells. Then, the next species is selected based on the evaluation of the energy function. There are four common selection techniques used in the GAs. We describe one of them that is called the *binary tournament* selection, which is as follows.

We set *a priori* the probability p_s of selection. The higher p_s , the more likely it is that the GA will eventually yield the true optimal solution. One selects a value of p_s that is high enough to ensure a reasonably accurate solution, but not too high to make the computations prohibitively expensive. One then picks at random two of the species from the population. A random number r , uniformly distributed in $(0,1)$, is generated. If $r < p_s$, one selects the species that resulted in a smaller energy. Otherwise, the one with a larger energy is picked. Another pair of species is then selected from the population and the same procedure is repeated. Each pair of the selected species constitute a *mating pair*. Note that since the species are chosen at random, any species may be selected numerous times, which is why the population of the selected species generally results in a set with lower energies. Repeating this procedure a large number of times finally yields a set of mating pairs, the size of which is half the population's size. Each mating pair can generate two offsprings during the crossover step. This completes the selection process.

Crossover is achieved when a mating pair produces two offsprings. To do so, the values of the parameters to be optimized are first converted to binary numbers involving zero and one. Similar to the selection, there are several types of crossover methods that one may utilize. One may, for example, utilize *uniform crossover*, since it has been shown to encourage the exploration of a robust search domain or population. For each parameter value in its binary representation, the GA selects a zero and one at random and exchanges their places in the parameter's binary representation. The procedure is then repeated a number of times to generate a new binary representation of that parameter of the offspring. For example, suppose that two of the parameters to be optimized are represented by 01110011010 and 10101100101. Then, one generates, by a uniform crossover, the offsprings 01110100101 and 10101011010. This procedure is repeated for all the parameters in all the mating pairs, resulting at the end in the new species or offsprings (which explains why using a large population of the species is very useful). The crossover step is then completed.

Mutation: After the offsprings are generated, it is possible that they exhibit some kind of "defect" which exhibits itself by resulting in an energy of larger numerical value. Such a defect in the GAs is known as mutation. The mutation probability is selected from a Gaussian distribution with zero mean and a given variance (e.g., 0.1). All the binary representations of the parameters are converted back to real numbers, hence generating the new population of the species. Numerical simulation of the phenomenon for which the data are available is then carried out using the new population of the offsprings or species, and the energies are computed for all the species based on the newly-calculated properties for which the data are available. This completes one generation of the computations. Then, the entire procedure, consisting of selection, crossover, mutation, and eliticism (see below),

is repeated again, resulting in the second generation. Enough generations are produced until the true optimal solution (in the sense that the minimum of the energy for a species is its true global minimum, not a local one) is obtained.

Eliticism: In order to speed up the computations and ensure that the true global minimum of the energy will eventually be reached, one also uses *eliticism* that, for every generation, selects those species with the energies of the lowest values and inserts them into the next generation.

Let us point out that the efficiency and performance of any GA depend on the numerical values of its parameters, ranging from the population size, to the selection, crossover, and mutation probabilities. This is a virtue of any GA since, by selecting proper (optimal) values of such parameters, the method's efficiency can be greatly increased, while lowering its computational cost.

7.5.4.2 Reconstruction Based on Flow and Seismic Data

Hamzehpour and Sahimi (2006b) and Hamzehpour *et al.* (2007) developed a reconstruction method for the FS porous media that utilized three distinct types of data.

1. Flow data that provide insight into the *connectivity* of the permeable regions of a porous medium. Hamzehpour *et al.* (2007) utilized data for the time-dependent fluid pressure at a production well.
2. Seismic data that provide insight into *large-scale* heterogeneities of a porous medium. Hamzehpour *et al.* (2007) utilized the arrival times of seismic waves at particular points in a porous medium.
3. Porosity logs (or any other type of log) that provide information about the *local* heterogeneities of a porous medium.

Using the SA method for minimizing the energy of the system, Hamzehpour and Sahimi (2006b) and Hamzehpour *et al.* (2007) showed that their reconstruction method produces models of the FS porous media that provide accurate information for those properties of porous media for which no data are available. In particular, Hamzehpour *et al.* (2007) showed that the reconstructed model provides accurate predictions for two-phase flow in porous media, which is usually very difficult to do, without using any data for the two-phase flow.

The crucial aspect of the reconstruction model of Hamzehpour *et al.* (2007) was that they used fluid flow data. Such data are of fundamental importance to not only single-phase flows, but, more importantly, to two-phase flows. Without such data and given that it is very difficult to obtain other types of data that provide information on the connectivity of high-permeability regions of a FS porous medium, no reconstruction method can be expected to result in realistic models of porous media that can provide accurate predictions for flow and transport processes in such media.

8

Models of Fractures and Fractured Porous Media

Introduction

In Chapters 4–6, we described several characteristics of porous media as well as those of fractures and fracture networks, and their experimental measurements. Chapter 7 described various models of unfractured porous media. In the present chapter, we describe and discuss models of a single fracture, a network of interconnected fractures, and fractured porous media. A study of the extensive literature on the subject indicates that there are several classes of models of fracture networks and fractured rock. Each has its own strengths as well as weaknesses. For example, the classical multiporosity models of fractured porous media that were proposed in the early 1960s have been heavily utilized in the oil industry, but have proven to be inadequate for modeling several types of fractured porous media. There exists a class of fracture network models that are, in essence extensions of the network models of porous media described in Chapter 7, and to some extent resemble the percolation networks described in Chapter 3.

8.1

Models of a Single Fracture

Similar to the models of unfractured porous media, the simplest model of a single fracture is the space between two parallel, infinite flat planes (Snow, 1969). The model has been used simply because slow (laminar) fluid flow in such a geometry can be solved analytically. However, the model has two weaknesses.

1. It cannot take into account the effect of internal surface roughness of fractures that was described in Chapter 6. One can, however, superimpose the surface roughness on the two flat plates (Gutfraind and Hansen, 1995; Zhang *et al.*, 1996; Drazer and Koplik, 2000, 2001; Madadi and Sahimi, 2003, 2011; Madadi *et al.*, 2003), resulting in a more realistic model of a single fracture (see also below).
2. If the mechanical properties of a fracture are important to modeling of fluid flow through it, the model is too simple to take such properties into account.

In an attempt to take into account the variations in the aperture of a fracture due to surface roughness, more sophisticated models began to emerge. Muradlidi (1990) and Zimmerman *et al.* (1992) used the two parallel flat plates model, but added obstacles to the space between them in order to study their effect on fluid flow. Another and perhaps more realistic way of modeling flow in a single fracture is by recognizing that due to the rough internal surface of the fractures, the local aperture is a spatially-varying property. It has also been observed that, under stress, the flow of a fluid through a single fracture is usually along certain channels. Thus, one way of modeling a single fracture is to map its internal morphology obtained through the characterization of the fracture onto an equivalent network of channels, through which a fluid flows. The effective permeability of the channels is, of course, related to the local apertures that are spatially correlated. Motivated by such considerations, Neuzil and Tracy (1981), Tsang and Witherspoon (1981), and Moreno *et al.* (1985) represented a single fracture by a two-dimensional (2D) network of bonds, the apertures of which were distributed randomly according to a log-normal distribution in one direction, but assumed to be constant in the second direction that was the direction of the applied pressure, hence resulting in rectilinear streamlines. In particular, Tsang and Witherspoon (1981) used experimental data for the apertures.

Another class of models of a single fracture consists of 2D networks in which stochastically-generated aperture fields are used, without taking into account the mean surface deviation. Moreno *et al.* (1988) and Tsang and Tsang (1989), for example, generated the apertures according to a log-normal distribution with exponentially decaying correlations, with the latter group using anisotropic correlations; see also Nolte *et al.* (1989). Zimmerman *et al.* (1991) and Hakami *et al.* (1995) modeled a fracture by a channel with undulated walls. The former group assumed the flow to be parallel to the undulations wavevector, hence resulting in non-rectilinear flow lines, but neglected flow channelization. The latter group shifted the undulated walls by a shear displacement oblique with respect to the undulations wavevectors, resulting in elongated contact areas and anisotropic aperture variograms in agreement with experimental data.

Assuming that the surface of fractures is self-affine, another class of models of a single fracture may be developed which probably represents the most realistic. The origin of such models goes back to the early work of Patir and Cheng (1978) in a context unrelated to flow in a single fracture. They were interested, instead, in the effect of surface roughness on flow in pipes with lubrication. In such models, the fracture is initially represented by a 3D channel between two parallel flat surfaces. One then generates the self-affine roughness profile and superimposes it on top of the flat surfaces. To generate the roughness profile, one may use a 2D (1D) fractional Brownian motion for a 3D (2D) fracture, described in Chapters 5 and 6. The local apertures δ vary spatially within the fractures. By translating the upper and lower surfaces normal to the mean plane, one may vary the distance between the upper and lower surfaces and obtain fractures with a range of apertures. If a single fracture is modeled this way, the local apertures need not be estimated

explicitly, as the governing equations for flow or a transport process through the fracture will be directly discretized and solved numerically.

Such a model has been utilized widely. Brown (1987b, 1995), Wang *et al.* (1988), Unger and Mase (1993), Plouraboué *et al.* (1995), Gutfraind and Hansen (1995), Gutfraind *et al.* (1995), Zhang *et al.* (1996), Drazer and Koplik (2000, 2001), Mourzenko *et al.* (1995, 1996, 1999, 2002), Madadi and Sahimi (2003, 2011), and Madadi *et al.* (2003) all generated rough fracture surfaces using one of the techniques described in Chapter 5 for the generation of rough surfaces. Each of the groups studied one or more aspects of fracture models and fluid flow through them. For example, Brown (1987b) allowed the mean separation of the two surfaces to vary, but ignored overlaps. Wang *et al.* (1988) and Plouraboué *et al.* (1998) generated identical rough surfaces with shear displacement in order to study the correlations of the apertures. Brown (1995) developed a model in which the fracture surfaces matched at long, but not short, wavelengths. Gutfraind and Hansen (1995), Gutfraind *et al.* (1995), Zhang *et al.* (1996), Drazer and Koplik (2000), Mourzenko *et al.* (1995, 1996, 1999, 2002), and Madadi *et al.* (2003) studied flow and conduction in a single fracture.

8.2

Models of Fracture Networks

The idea that a fractured rock can be represented by a network of interconnected and finite fractures is intuitively clear and appealing, and is also supported by the experimental data described in Chapter 5. Thus, representing fractured rock by a discrete network has a relatively long history. Romm (1966), Parsons (1966), Snow (1969), Caldwell (1972), and Wilson and Witherspoon (1975) appear to be among the first to have used such an approach. They used electrical analog models to study flow through a network of fractures. In particular, Parsons (1966) used square and triangular networks of resistors in which each bond (or resistor) represented a finite fracture. However, his model had the drawback that the current in each resistor was assumed to be proportional to the width of the conductor, whereas the flow rate in a fracture is proportional to some power of the fracture's aperture that, as described in Chapter 6, can be as high as six (see also Tsang and Witherspoon, 1981). Snow (1969) used a 3D model in which the fractures were idealized as infinitely long and parallel ducts. As such, his model was in the same spirit as the bundle of parallel capillary tubes described in Chapter 7. Similar models were also developed by Castillo *et al.* (1972) and Krizek *et al.* (1972). We consider 2D and 3D fracture networks separately, and study their properties. However, before doing so, a few key concepts are introduced.

8.2.1

Excluded Area and Volume

An important concept in the modeling of a fracture network is the excluded volume. Balberg *et al.* (1984a) defined the excluded volume of an object as the volume that surrounds it, in which the center of another object must be in order for the two object to be connected, with a similar definition for 2D objects. Consider the case of identical objects, and keep in mind that such concepts are meaningful only if the objects' locations are uniformly distributed in space. We summarize the excluded volume $\mathcal{V}_e$ for some simple cases:

$$\mathcal{V}_e = \begin{cases} \frac{2}{\pi} \ell^2 & \text{line segments in a plane ,} \\ 4S & \text{disks in a plane ,} \\ \pi^2 R^3 & \text{disks in 3D space ,} \\ 8V & \text{spheres ,} \\ \frac{1}{2} \pi^2 (e_1 l_1^2 l_2 + e_2 l_1 l_2^2) & \text{ellipses in 3D space ,} \end{cases} \quad (8.1)$$

where ℓ is the line segment's length, R and S are the disk's radius and area, respectively, and V is the sphere's volume. The first equation is for the case in which the line segments are randomly and uniformly oriented, while the second equation is valid if all the disks have identical orientations. The third equation, derived by Charlaix *et al.* (1984), is for the case in which the disks are randomly oriented in space. The last expression was derived by de Dreuzy *et al.* (2000) for ellipses of eccentricities e_1 and e_2 , and of major axis lengths l_1 and l_2 . If the length of the line segments, or the radius of the disks, is distributed according to some statistical distributions, then

$$\mathcal{V}_e = \begin{cases} \frac{2}{\pi} \langle \ell^2 \rangle & \text{line segments in a plane ,} \\ \frac{\pi^2}{8} \langle R^2 \rangle \langle R \rangle & \text{disks in 3D space .} \end{cases} \quad (8.2)$$

Balberg *et al.* (1984a) suggested another type of averaging in place of what is indicated by Eq. (8.2):

$$\mathcal{V}_e = \begin{cases} \frac{2}{\pi} \langle \ell^2 \rangle & \text{line segments in a plane ,} \\ \frac{\pi^2}{8} \langle R^3 \rangle & \text{disks in 3D space .} \end{cases} \quad (8.3)$$

The above results were for simple objects. Ishihara (1950) developed a general method for randomly oriented objects. Consider 3D convex objects A and B with surfaces S_A and S_B , volumes V_A and V_B , and surface averaged mean radius of curvatures $\mathcal{R}_A$ and $\mathcal{R}_B$. Then, the *mutual excluded volume* $\mathcal{V}_{AB}$ is given by

$$\mathcal{V}_{AB} = V_A + V_B + (S_A \mathcal{R}_A + S_B \mathcal{R}_B) . \quad (8.4)$$

Equation (8.4) may be averaged over the distributions of the objects' shapes and sizes. It yields, for example, the result for spheres given by Eq. (8.1). If the objects

are flat and convex, one obtains

$$\mathcal{V}_{AB} = \frac{1}{4} (S_A P_A + S_B P_B) , \quad (8.5)$$

where P_A is the perimeter of object A . Thus, if Eq. (8.5) is averaged over the size distribution of objects with identical shapes, one obtains

$$\mathcal{V}_{AB} = \frac{1}{2} \langle S \rangle \langle P \rangle , \quad (8.6)$$

which is a general result. A different expression, $\mathcal{V}_e = \frac{1}{2} \langle S P \rangle$, has also been suggested and sometimes used.

8.2.2

Two-Dimensional Models

We should point out at the outset that natural fractures are not generally planar. This was discussed in some detail by Gertsch (1995). However, many models of fracture networks have been developed (see below) in which the fractures are represented by planar objects. Such models, which represent simplifications of the actual fracture networks of rock, have, nevertheless, proven to be useful and realistic.

Over the past three decades, many research groups have developed 2D models of networks of fractures of finite extent. In such models (Dienes, 1980; Long *et al.*, 1982; Engleman *et al.*, 1983; Schwartz *et al.*, 1983; Smith and Schwartz, 1984; Charlaix *et al.*, 1984, 1987a; Endo *et al.*, 1984; Robinson, 1984a,b; Long and Witherspoon, 1985; Ross, 1986; Long and Billaux, 1987; Shimo and Long, 1987; Guéguen and Dienes, 1989; Hestir and Long, 1990; Robinson and Gale, 1990; Mukhopadhyay and Sahimi, 1992) the fractures are represented by 1D finite line segments, to which effective permeability is attributed. Thus, such fracture networks are similar to 2D networks of pores already described in Chapter 7. They can even be a reasonable representation of a 3D network of interconnected fractures if most of the hydraulic conductivity is in the intersections *between* the fractures, or if fluid flow is channelized in the fractures. Flow channeling in fractures does happen sometimes, in which case, a 2D model of network of fractures may be useful. On the other hand, Madadi and Sahimi (2003, 2011) constructed a 2D fracture network in which each fracture had an internal rough, self-affine surface superimposed on two parallel flat planes (see Section 8.1) and studied its fluid flow and transport properties.

In the 2D models, one distributes the fractures at random in a plane. One of the simplest models is the Poisson model that was first used by Long *et al.* (1982). In their model, one selects in a square block of size $L \times L$ the x - and y -coordinates for a specified number of lines or fractures centers from a uniform distribution in $(0, L)$. Once the centers' coordinates are selected, the orientation of the lines are also selected from a given distribution. Then, the lines are assigned randomly-selected lengths and hydraulic conductances. If the fractures cross the boundary of the system, they are truncated, but no truncation is done inside the $L \times L$ box.

Figure 8.1 shows a typical network obtained by the Poisson method. As such, such models are practically identical with the percolation networks of sticks studied by Balberg *et al.* (1991, 1984b) and Balberg and Bienbaum (1983, 1985) that were mentioned in the discussion of continuum percolation in Chapter 3. In particular, Balberg and Bienbaum (1983) studied 2D anisotropic networks of conducting sticks that may be a more realistic representation of 2D naturally-fractured porous media that are usually anisotropic. Moreover, Balberg *et al.* (1984b) considered a 3D fracture network in which the fractures were finite cylinders of length L and radius r , and studied the dependence of the network's percolation threshold on the aspect ratio L/r and the macroscopic anisotropy.

Such networks of fractures do have a major difference with the percolation networks. In a percolation network, an upper bound to all the properties corresponds to their values at $p = 1$, where p is the fraction of conducting bonds (see Chapter 3). All the quantities of interest are then normalized with respect to their values at $p = 1$ because, otherwise, any property of the network would depend on its linear size. This is true about *any* network model. On the other hand, there is no theoretical end to the degree of fracturing. If one adds one more fracture to the network, its permeability or hydraulic conductivity increases infinitum. Thus, one cannot determine how “filled” a fracture network is. As Hestir and Long (1990) pointed out, one can study networks in which λ_ℓ , the average frequency of fractures intersecting a sample line, is held constant. Since any fracture network can be rescaled to a given constant λ_ℓ if it is held fixed, then the permeability of the network has its maximum value when all the fractures are *infinitely long*, which is the limit that Snow (1969) studied. Thus, if λ_ℓ is held fixed, Snow's analytical results for the permeability of a fracture network is equivalent to $p = 1$ limit of percolation networks.

The next issue to resolve is how to relate the parameters of fracture networks to those of percolation networks, such as p and $\langle Z \rangle$, the average coordination number

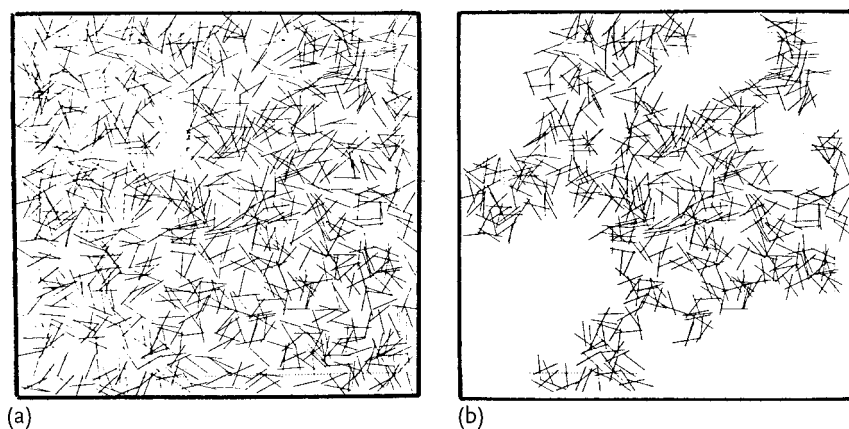


Figure 8.1 A 2D random fracture network (a) and its sample-spanning portion (b) (after Robinson, 1984b).

or connectivity of the network. Robinson (1984a,b) and Charlaix *et al.* (1987a) used the average number of intersections per fracture, ζ , as the measure of the connectivity (see below). Suppose that the average fracture length is $\langle \ell \rangle$, the orientation distribution of the fractures is $g(\theta)$, and the density of the fractures' center is λ_A . It is not too difficult to show that $\lambda_\ell = \langle \ell \rangle \lambda_A$, and

$$\zeta = \lambda_\ell \langle \ell \rangle \Theta, \quad (8.7)$$

with

$$\Theta = \int_0^\pi \int_0^\pi \sin |\theta - \theta_0| g(\theta) g(\theta_0) d\theta d\theta_0. \quad (8.8)$$

For example, if the orientations of the fractures are uniformly distributed, $g(\theta) = 1/\pi$ and $\Theta = 2/\pi$. Now, for every ζ , there is a $p_f(\zeta)$, the analog of p in the percolation networks. For example, in the fracture network of Engelman *et al.* (1983), one has $p_f(\zeta) = 1 - \exp(-\zeta)$. Therefore, a critical value of ζ , ζ_c may also be defined and, thus, all the results of percolation theory described in Chapter 3 may be written in term of ζ if we replace p everywhere in the percolation theoretical results with $p_f(\zeta) = 1 - \exp(-\zeta)$.

As the second example, consider Robinson (1984a,b)'s model that was analyzed by Hestir and Long (1990). In his model, $p_f(\zeta)$ is the average fraction of a fracture that is available for flow. Now, consider a fracture of length ℓ with $n(\ell)$ intersections. If the fracture length is constant, then $n(\ell)$ will be a Poisson process. The average fraction of a fracture available for flow, that is, the fraction that is between the two end sites separated by the distance ℓ , is $[n(\ell) - 1]/[n(\ell) + 1]$. Therefore, if P_n is the probability that $n(\ell) = n$, one simply has $P_n = (n!)^{-1} \zeta^n \exp(-\zeta)$, and

$$p_f(\zeta) = \sum_{n=2}^{\infty} \frac{n-1}{n+1} P_n = \left(1 + \frac{2}{\zeta}\right) [1 + \exp(-\zeta)] - \frac{4}{\zeta}. \quad (8.9)$$

Once again, all the results of percolation theory may be copied for Robinson's model by replacing p everywhere in percolation theory with p_f given by Eq. (8.9). Hestir and Long (1990) worked out several other examples that related p_f (or p) to ζ . They also considered the case in which the fracture lengths were not constant, but were distributed according to a given statistical distribution. Developing the relation between p and p_f is important to computing the effective flow and transport properties of fracture networks. For example, Hestir and Long (1990) used the relation in the effective-medium approximation (see Chapters 9 and 10) for the effective conductivity and permeability of random networks as well as the scaling theory of the permeability near the percolation threshold p_c (see Chapter 3) in order to predict the hydraulic conductivity of 2D fracture networks, which necessitates relating p (or p_f) to ζ .

Long and Billaux (1987) developed a 2D network model of fractures by incorporating field data into the model. The network was generated subregion by sub-

region, such that the properties of each subregion were predicted through geostatistics. The region in which the data were collected was divided into statistically-homogeneous subregions. The fractures were divided into five sets based on their tectonic history. It was observed that in each set, the fractures were spaced close together and had similar orientations. The information was incorporated into the network model. Another piece of information that was used in the simulation was the aperture distribution. The criterion for accepting a model was its ability for reproducing the measured permeability of the subregion. After all subregions were created, they were joined together to recreate the entire region. When the model was applied to Fanay-Augères, a uranium mine in France, it was found that, macroscopically, the region was barely connected. Only 0.1% of the fractures contributed to the permeability of the region. This implied that (1) the system may have the topology of a percolation network very close to the percolation threshold p_c (if not at p_c), and (2) the fracture network was a fractal object over the scale of the observation. Both findings support the ideas of Sahimi *et al.* (1993, see Chapter 6), further discussed below, that at large length scales, the fracture network of rock should be similar to a percolation network. The results also appear to be typical of many fractured porous media, and indicate the significance of percolation concepts to modeling of such fracture networks.

The applicability of 2D networks of fractures to modeling naturally-fractured porous media is limited. One main reason for this is that a 2D model cannot realistically describe the fracture network connectivity in a 3D space because the fractures that do not connect in a planar cut may connect in the third dimension. Moreover, whenever 2D data are used with a 3D model, one may have a non-uniqueness problem in the sense that many distinct 3D models may account for the same 2D data.

8.2.3

Three-Dimensional Models

Bour and Davy (1997, 1998) extended the 2D networks of sticks or line segments to 3D. Other than their work, in the 3D models, the fractures have been represented by either discs of finite radius (Warburton, 1980a; Long *et al.*, 1985; Shapiro and Andersson, 1983; Andersson and Dverstorp, 1987; Charlaix *et al.*, 1987a; Liggett and Medina, 1982; Billaux *et al.*, 1989; Tsang *et al.*, 1988; Piggott and Elsworth, 1989; Cacas *et al.*, 1990a,b; Nordqvist *et al.*, 1992; Piggott, 1997), or by flat planes of finite dimensions (Wilke *et al.*, 1985; Berkowitz, 1995). A third class of models in which the fractures are represented by regular or irregular polygons has been developed by Adler, Thovert, and co-workers, and will be described separately. Chilés (1988), on the other hand, described the use of fractal and geostatistical methods in the development of models of a fracture network.

There is experimental evidence that 3D fractures are either roughly elliptical or discs-shaped (Pollard, 1976). In some cases, compared with an actual porous medium that we wish to study, the fractures are very long, and thus they can be considered as essentially infinite. In this case, a simple stochastic model based on the

Poisson distribution, which is an extension of the 2D networks described earlier, may suffice. The Poisson process can also be used for generating clusters of fractures that are concentrated in a given subregion. The center of each subregion is generated by the Poisson process and, then, around each center, a cluster of fractures are generated. Similar to conditional simulation of flow in large-scale porous media in described in Chapter 7, any realistic model of fracture networks should also honor the available experimental data. Thus, if, for example, the conditioning of the model (to force it to honor the data) ought to be done with respect to the observed fracture density, one first generates the fracture network, rejects all the fractures that intersect the surveyed lines or surfaces, and then adds the actual fractures that have been deduced through the analysis of field or laboratory data. This is particularly simple with a Poisson model because in this model, each fracture is generated independently of all the others.

There have been some attempts to use a 3D fracture network for simulating the hydraulic behavior of a given field and match the measured data. Billaux *et al.* (1989) extended the model of Long and Billaux (1987), described above, to three dimensions. The fractures were represented as disks placed randomly in space. The diameter of each disc was selected independently from a probability distribution that was assumed to be log-normal. To locate the fractures in the space, a point process called the *parent-daughter process* was used. In this method, one uses a Poisson process to place a cloud of points (or daughters) around each Poisson point called a parent or seed. The number of points in each cloud is a Poisson random variable, and each point is placed in a given cloud independently of all the other points. The motivation for doing so is the fact that the experimental data indicate that fractures of rock often occur in swarms.

Figure 8.2 shows a typical swarm of disk-like fractures. As in the case of the 2D model (Long and Billaux, 1987), the fractures were divided into five different sets. The orientation of the discs in each set was characterized as a fluctuation about

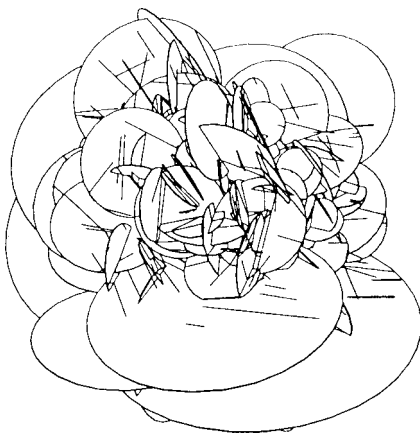


Figure 8.2 A 3D network of disk-like fractures (after Billaux *et al.*, 1989).

the mean orientation for the set. The fluctuations had a spatial structure that could be simulated with geostatistics. After each set was generated, a model of a given fractured field was created by putting together all of the sets. Using the model, the hydraulic and transport properties of the fractured rock were simulated. In a work somewhat similar to that of Billaux *et al.* (1989) and Dverstorp and Andersson (1989) used a 3D network of disc-like fractures and showed that the model can be calibrated by one set of data, and then be used to predict and match another set of data. These will be further described in Chapter 11.

8.2.4

Fracture Networks of Convex Polygons

Models that use flat planes to represent fractures have been significantly refined and extended by Adler, Thovert and their group (Huseby *et al.*, 1997, 2001; Koudina *et al.*, 1998). In their model, the fractures are represented by convex polygons. An example is shown in Figure 8.3. To describe the model, we consider the model of Koudina *et al.* (1998). Although, as discussed above, the fractures can, in principle, have any shape, size, permeability, or spatial distribution, Koudina *et al.* (1998) considered a particular model in which plane polygonal fractures are inserted in a simulation cell of linear dimension L . The fracture's normal vectors are randomly and isotropically distributed, with their center following a Poisson distribution. The contour of a fracture is a convex polygon inscribed in a circle of radius R (see Figure 8.4). The location of a fracture is given in terms of the center of the circle that surrounds it.

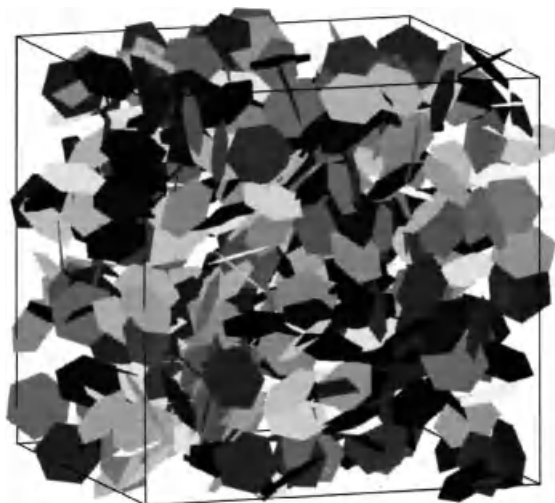


Figure 8.3 A 3D network of fractures represented by intersecting polygons (after Koudina *et al.*, 1998).

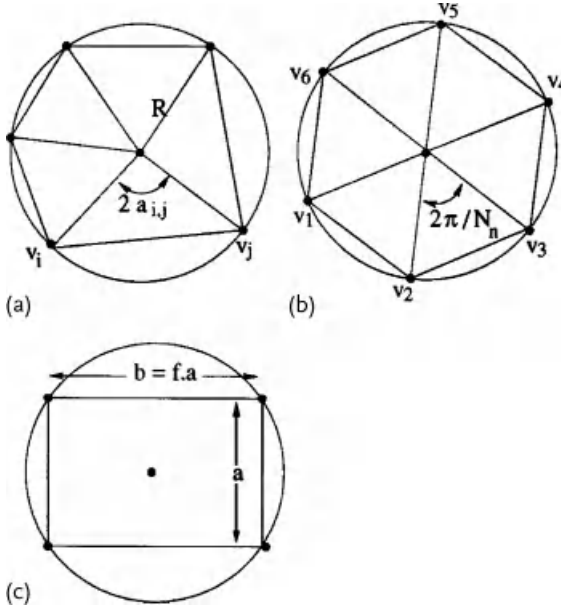


Figure 8.4 Geometry of fractures represented by regular or irregular polygons and inscribed in a circle of radius R . (a) Irregular; (b) regular hexagons, and (c) rectangular fractures (after Koudina *et al.*, 1998).

The vertices of the polygon are evenly (for a regular polygon) or randomly (for irregular polygons) distributed on the perimeter of the circle. The number of vertices of each fracture is selected randomly and uniformly in the interval $[3, N_{\max}]$. The vertices are distributed on the disk's contour with angles that are distributed uniformly in $[0, 2\pi]$. The orientation of the polygon within its plane is random, distributed uniformly. Hence, the fracture network is completely characterized by the fractures' shape, the ratio L/R , and the number of fractures per unit volume. As described earlier, one can also take into account the effect of the excluded volume $\mathcal{V}_e$ of a fracture defined as the surrounding volume into which the center of another fracture cannot enter if overlap between the objects is to be avoided.

The fractures' intersections are then determined, with the intersection lines characterized by their end-point coordinates. Also identified are the triple points where three fractures intersect one another. If the system is large enough, then, at the macroscopic scale, the fracture network can be considered as homogeneous and, therefore, one may use periodic boundary conditions. One also must check whether the generated fracture network is sample-spanning to ensure the existence of macroscopic flow and transport. One must also identify the connected part of the fracture network. This is done using an algorithm developed by Thovert *et al.* (1993). Identifying the connected, sample-spanning part of a fracture network is important, as it simplifies the computations of flow and transport properties. On the other hand, if the matrix contributes significantly to the flow or transport

processes, identifying both the connected and disconnected part of the fracture network is important.

In order to carry out flow and/or transport calculations in the network, one must discretize the fracture network. Given the shape of the fractures, the most suitable discretization is unstructured triangulation that, however, must satisfy several constraints: The vertices of all the polygons, intersection end-points, and triple points must coincide with vertices of the triangular mesh. In addition, the border lines of the original polygons and fracture intersections must coincide with triangle edges. Moreover, the triangulation of two intersecting fractures must match along their intersection line.

To start the triangulation process, one first identifies the fractures that belong to the sample-spanning clusters so that the isolated fractures can be discarded and not triangulated. However, to better understand the process of triangulation, one must first consider the case of a single isolated fracture in the form of a convex polygon, which we refer to as Ω_0 , with its contour being $\partial\Omega_0$ (see Figure 8.5). Suppose that Ω is that part of Ω_0 that has not been triangulated yet, with its contour being $\partial\Omega$ (see Figure 8.5a), which represents the advancing front (the front that separates the triangulated zone from the untriangulated segment). The triangulation involves two basic operations which are the junction of two second-neighbor points of the front (see Figure 8.5c,e) if the resulting triangle is acceptable, and insertion of a new point within Ω , which would be a new triangle when the inserted point is connected with the two facing points of $\partial\Omega$ (see Figure 8.5d). At each step of the operation, the front $\partial\Omega$ is updated by removal or insertion of points. The operation stops when $\partial\Omega$ is reduced to an acceptable triangle.

The more complex case involves a fracture that intersects other fractures, an example of which is shown in Figure 8.6. In this case, the intersection line may cross the boundary of the fracture once or twice, hence splitting it into two disconnected zones. Alternatively, the intersection line may lie entirely within the fracture. One may also have a situation in which many intersection lines, themselves intersecting one another, may enclose internal subdomains with or without connections with the outer contour of the fracture. There are two ways to triangulate such a fracture. In one method, after identifying each subdomain, it is treated as an independent polygon (and presumably an irregular one due to the random positions of the intersecting lines), and then its triangulation is carried out separately.

An alternative method, developed by Koudina *et al.* (1998), is as follows. As the first step, all the internal features are connected to the outer boundary of the fracture (see Figure 8.6b). They are also split into smaller segments with a length smaller than a given value δ_m . In the next step, a cycle $\partial\Omega_0$, which describes the fracture boundary and all of its internal features, is constructed and treated as the outer contour of a single simply-connected (non-convex) polygon (see Figure 8.6c). The splitted cycle $\partial\Omega_0$ contains the first set of points to which the advancing front technique is applied. The same rule, using the same δ_m , is applied to all the fractures, and a unique splitting is used for the intersection line of two fractures. Moreover, during

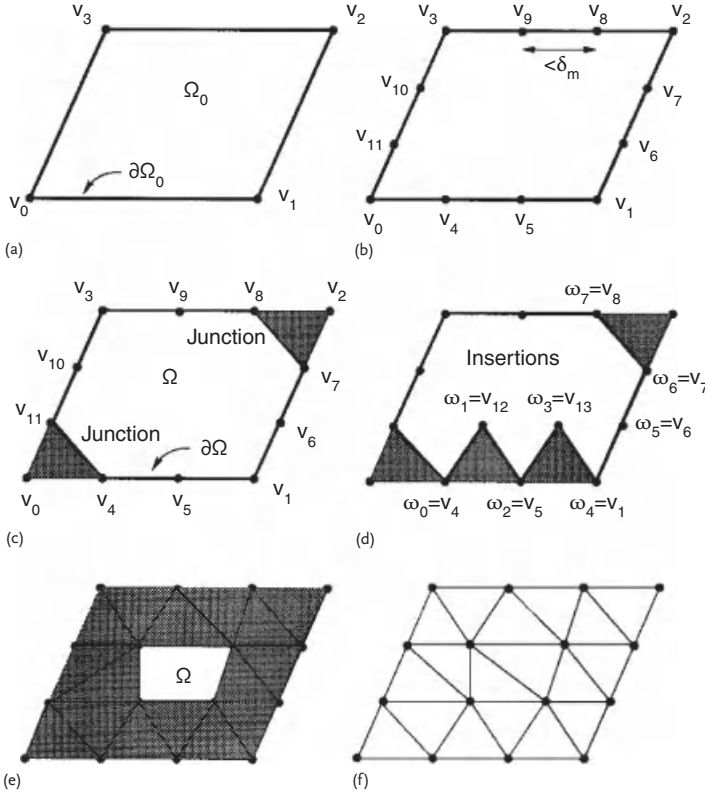


Figure 8.5 The steps of triangulation of a simple domain by an advancing front. (a) The initial domain Ω_0 contains four vertices, which is then partitioned into four segments

shorter than δ_m in (b), which then has 12 vertices. The final triangulated system is shown in (f), with the figures in (c)–(e) showing the intermediate steps (after Koudina *et al.*, 1998).

the insertion operation in which a new point $v_{N_{p+1}}$ from a segment (ω_i, ω_{i+1}) of $\partial\Omega$ is inserted into the domain to create a new fracture (see Figure 8.6d), $v_{N_{p+1}}$ is set such that

$$\|\omega_i v_{N_{p+1}}\| = \|\omega_{i+1} v_{N_{p+1}}\| = \delta_I, \quad (8.10)$$

where, although initially, $\delta_I = \delta_m$, but δ_I is modified as the process advances. Another constraint imposed on the system ensures that the edges $(\omega_i, v_{N_{p+1}})$ and $(\omega_{i+1}, v_{N_{p+1}})$ do not cross any existing edge. The intermediate stages of this process are shown in Figure 8.6d–f.

Regardless of the triangulation method used, one finally obtains a collection of triangles that are defined by their vertices for each fracture. Then, the entire fracture network consists of a union of all the fractures and their triangles. As mentioned above, the triangulation of the intersecting fractures match along their in-

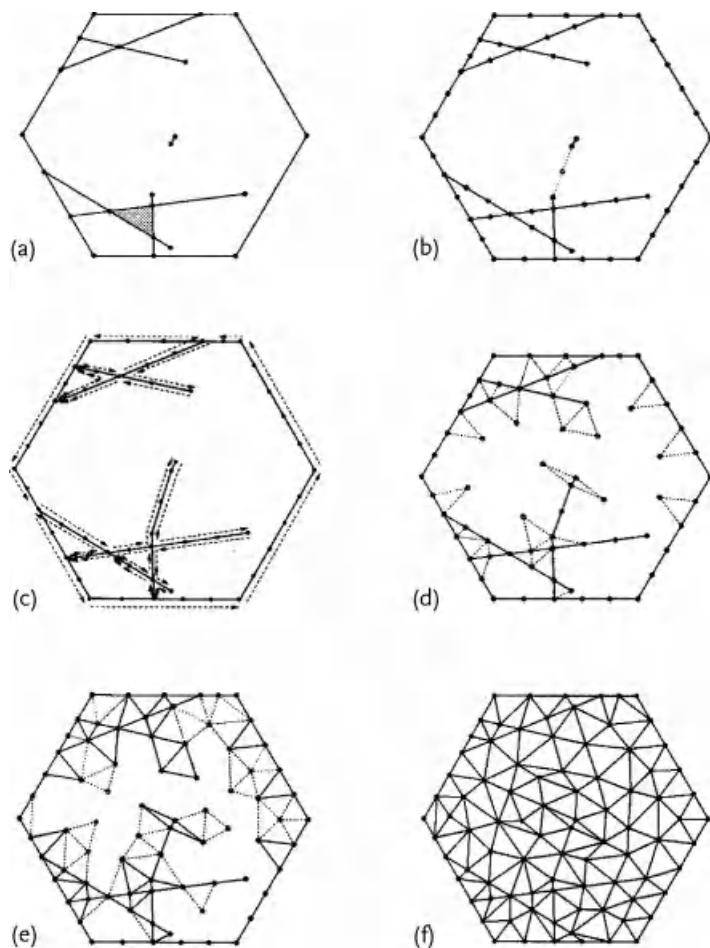


Figure 8.6 Same as in Figure 8.5, but for a fracture that has been intersected by other fractures. The shaded area in (a) indicates a subdomain (after Koudina *et al.*, 1998).

tersection line since the procedure applied splits the fracture in its two embedding planes. Each triangle belongs to a fracture, but its edges may be the intersection of two fractures, hence belonging to both. Since isolated fractures that do not belong to the sample-spanning cluster are not triangulated, all the generated triangles play a role in the simulation of flow and transport in the fracture network. Figure 8.7 shows the various triangulations that one obtains by varying the ratio δ_m/R . For more details, also see Adler and Thovert (1999).

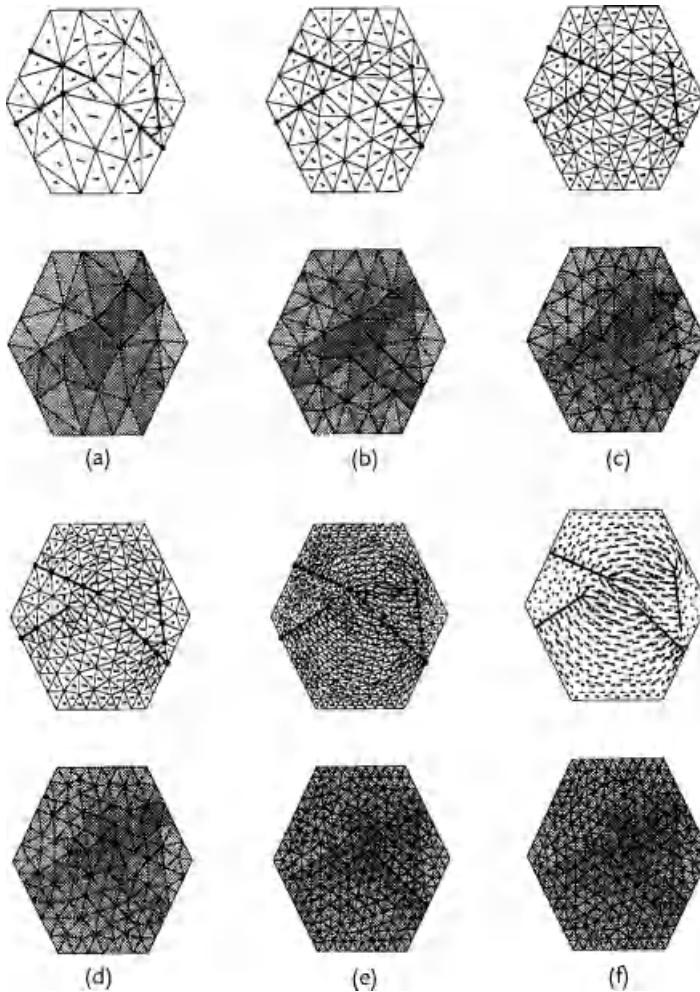


Figure 8.7 Various triangulation of a fracture with, respectively, $\delta_m/R = 1/2$ $1/3$ $1/4$ $1/5$ and $1/8$ shown in (a)–(f). Larger black circles show the grid points in common with other fractures that have intersected the fracture

shown. For each panel, a similar figure at the top shows the direction of the fluid flow in which the size of the arrow is proportional to the fluid flux (after Koudina *et al.*, 1998).

8.2.5

The Dual Permeability Model

Models of fracture networks of the type described above are very useful for gaining a deeper understanding of flow and transport phenomena in fractured rock. The main disadvantage of such models is that if they are to be used for modeling of fractured rock at large scales (of the order of at least several hundreds of meters), the computations of even the simplest flow problem may become pro-

hibitively long. For this reason, a continuum representation of fracture networks would be desirable. However, in order not to be hampered by the same type of shortcomings that the classical models of fractured porous media suffer from (see below), one must first use network simulations to gain a better understanding of a given phenomenon at smaller length scales, and then identify an appropriate representative elementary volume (REV) for averaging the simulation results. Doing so enables one to transfer the smaller-scale results to a larger length scale, that is, the continuum model.

In doing so, one must assume that the connectivity properties of the smaller-scale network used in the simulations are representative of the much larger fracture network, the simulation of which is not feasible. As discussed below, this assumption is not always justified, but has been used in the past. For example, although the fracture network is sample-spanning and above its percolation threshold, it may be quite close to the percolation threshold, a situation that is encountered quite often in fractured rock. Thus, the correlation length of the network associated with percolation (see Chapter 3) is very large and, therefore, a REV, even if it can be defined, would be so large that averaging the results of the network simulations and transferring the results to larger length scales may become meaningless. Moreover, if the fracture network is a fractal object (see Chapter 6), the linear size of the REV must be larger than the length scale over which the network has fractal characteristics, and often such length scales are very large.

An alternative method was suggested by Clemo and Smith (1989). They proposed the use of a hierarchy of fractures, from the smallest ones to the largest. In this scheme, only those fractures that are deemed more important to flow are modeled individually. Clemo and Smith (1989) called such fractures *primary*. It should be clear from our discussion of percolation networks in Chapter 3 that the primary fractures must be part of the backbone of the fracture network, that is, the multiply-connected part of the network through which flow and transport take place. Moreover, such fractures have the largest apertures in the network. By this criterion then, the vast majority of the fractures are not as important to flow and transport through the network as the primary fractures.

Such fractures are, therefore, classified in groups and are represented by some effective properties. The groups are called network blocks. Together, the combination of the primary fractures and network blocks constitute what Clemo and Smith called the *dual permeability model*. However, this should not be confused with the classical double-porosity model to be described below since the dual permeability model is just a way of classifying the fractures according to their importance to flow and transport in the entire fracture network. Note that although the individual fractures that are grouped in the blocks are not as important as the primary ones, the overall contribution of the blocks is about the same as that of the primary fractures.

Figure 8.8 shows a typical fracture network and its equivalent dual permeability model. The effective properties of the blocks, for example, their hydraulic conductance and porosity, are calculated from those of the fractures that they contain. Simulation of flow and transport in such a network is much less time consuming than the original fracture network and will be described in Chapter 12.

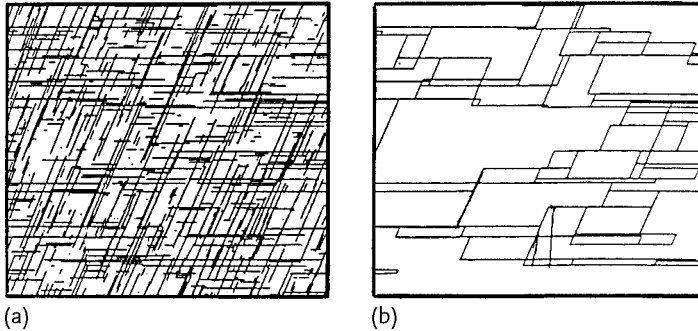


Figure 8.8 (a) A 2D fracture network and its (b) equivalent dual-permeability model (after Clemo and Smith, 1989).

8.3 Reconstruction Methods

We already described in Section 7.4 how optimization methods may be used for reconstructing models of disordered porous media. The same ideas and concepts may be utilized for developing models of fracture networks that honor a given set of data. We already described the models that were developed by Long and Billaux (1987), Billaux *et al.* (1989), and Dverstorp and Andersson (1989). A basic problem may, however, limit the usefulness of such models. The limitation is due to the fact that although there may be a large number of fractures in a field-scale porous medium, only a very small fraction or number of them usually contribute to the fluid flow and transport processes. That is, most of such fracture networks are at or near their percolation or connectivity threshold, may have fractal (scale-invariant) properties and, therefore, cannot be treated with the classical methods of analysis.

To overcome this difficulty, Long *et al.* (1991) and Davey Mauldon *et al.* (1993) used a simulated annealing (SA) method to reconstruct the structure of the fracture network in such a way that it honors the available data. In their work, the fracture network is represented by a 3D network of finite fractures. Then, the SA method is used to develop a network of connected fractures that accurately reproduces the experimental information about fractured porous medium. The most important issue to address is to determine how the fractures are connected so that the simulated flow and transport properties of the fracture network honor the data.

Similar to the reconstruction of unfractured porous media described in Chapter 7, the SA method may be used if one has (1) a set of possible configurations of the system; (2) a method for systematically changing the configurations; (3) an “energy” function to minimize, and (4) an annealing schedule of changing a temperature-like variable so that the system can reach its minimum “energy” – its ground state – at which the fracture network takes on a configuration that honors the existing experimental data.

For fluid flow in fractured porous media, the requirement (1) is well-defined: the set consists of networks of fractures in which some of the fractures allow fluid flow

to take place (they are “on”), whereas others are closed to flow or a transport process (they are “off”). Suppose that $\{C\}$ denotes the set of all the possible configurations of on and off fractures, similar to percolation networks of open and closed bonds. Requirement (3) is a method according to which a given configuration of the fracture network is varied. Suppose that one assigns a probability distribution function for randomly selecting a fracture in the network. Then, if the selected fracture is on, it is turned off and vice versa. The method generates a new configuration of the fracture network. One now defines the *neighborhood* N_C of C as the set of all the configurations that are very “close” to C or, in the language of Long *et al.* (1991), they are one step away from C .

Similar to the reconstruction of porous media based on the SA method, annealing the system means selecting a configuration of the fracture network from the set N_C and comparing it with C . In order to make the comparison precise, one needs to define an “energy” E , the third ingredient of the SA method. Similar to the discussions in Section 7.4, Long *et al.* (1991) defined E by

$$E = \sum_j |f(O_j) - f(S_j)|^\gamma, \quad (8.11)$$

where O_j and S_j are vectors of *observed* and *simulated* responses of the network, respectively, γ a constant, and $f(x)$ is a real monotonic function. The data could be hydrological, geophysical, or geological. A probability distribution of the configurations is then assumed to be expressible as the Gibbs’ distribution

$$p(C) = a \exp \left[\frac{E(C)}{T} \right], \quad (8.12)$$

where a is a renormalization constant that is very difficult to estimate because one must know the energy of *all* the configurations, which is impossible (but unnecessary, anyway), and T is a temperature-like variable to be defined below.

Because $p(C)$ is a Gibbs’ distribution, C , the current configuration of the fracture network may be modeled as a Markov random field, implying that the transition probability for moving from C to C' depends only on C and C' , and not on the previous configurations from the set $\{C\}$. Thus, the transition probability can change from configuration to configuration, but it does not depend on the previous configurations that were examined. Therefore, given C and N_C , the transition probability for moving from C to C' (given the current configuration C) is equal to the probability that one selects C' times the probability that the network would make the transition to a given configuration C' . Therefore,

$$p(C \rightarrow C' | C) = \begin{cases} 0 & C' \notin N_C, \\ P(C' | C) \times 1 & C' \in N_C, \\ P(C' | C) \exp\{[E(C) - E(C')]/T\} & C' \in N_C, \\ P(C' | C) \exp\{[E(C) - E(C')]/T\} & E(C') - E(C) > 0, \end{cases} \quad (8.13)$$

where for the last equation, we must have $C \neq C'$.

The final ingredient of the model is a schedule for the annealing, implying that as the annealing continues, one is less likely to keep those configurations of the fracture network that increase the energy E . The temperature schedule that Long *et al.* (1991) used was *ad hoc*, but effective. It is one that changes T in such a way that a number of iterations suffices to produce a fixed number of acceptable changes in the configuration of the fracture network. At the end of each iteration i , the temperature $T(i)$ is decreased using a geometric series

$$T(i+1) = T(i)R_i^i, \quad (8.14)$$

where $0 < R_i < 1$. Thus, one only needs to select the initial temperature, which is selected such that it is of the same order of magnitude as the energy difference between the first two configurations. Therefore, the energy difference between successive configurations remains (most of the time) between zero and one.

What does one do if, for example, quantitative information, for example, the range of possible responses, is available, but there are no actual measurements? Suppose, for example, that one wishes to predict the volume flow rate Q at a point in the network far from a point at which a data point is available, and has the insight that $a \leq Q \leq b$. In this case, each time a configuration of the fracture network is changed, the following steps are also taken:

1. the new point is added to the configuration and Q is calculated at that point.
2. A new energy function E' is calculated such that $E' = 0$, if the calculated flow rate $a \leq Q \leq b$; $E' = (Q - a)^\gamma$, if $Q < a$, and $E' = (Q - b)^\gamma$ if $Q > b$;
3. if $E(C') + E'(C') < E(C) + E'(C)$, then C' is kept. Otherwise, the usual annealing probability is used to keep or reject C' , and
4. the new point is removed and the process continues.

Long *et al.* (1991) showed that even if the initial configuration is a completely connected and dense network of open (“on”) fractures, the final configuration is usually a network that is *barely connected*, although it contains a very large number of disconnected fractures. This implies that the fracture network may be similar to the sample-spanning percolation cluster just above the percolation or connectivity threshold in agreement with the proposal of Sahimi *et al.* (1993), which was described and discussed in Chapter 6 and will also be discussed below.

The technique that we described has been used for reconstructing models of fractured porous media. For example, Datta-Gupta *et al.* (1995) utilized the method to reconstruct a model of fractured limestone formation. Day-Lewis *et al.* (2004) utilized a SA technique for identifying the fracture-zone geometry in the Mirror Lake Site in New Hampshire. The main problem with the reconstruction method is that for a large enough network, the computations become very time consuming and may not be feasible unless, of course, highly efficient strategies for example, massively-parallel computations, are utilized.

8.4

Synthetic Fractal Models

As described and discussed in Chapter 6, in many situations, fracturing of a porous medium is a product of a fragmentation process. The distribution of the fragment sizes is often a power law, a signature of a fractal system or process. A power-law distribution of the fragment sizes has been known since the 1940s when it was first discovered in grinding operations. Gilvarry (1964), for example, used an exponential distribution for describing repetitive fracturing. He assumed that imperfections, which are randomly distributed in rock, initiate fracturing. An important parameter of his analysis was $p_f(\ell)\delta\ell$, the probability that a fragment with a size between ℓ and $\ell + \delta\ell$ can be fractured. Turcotte (1986) was probably the first to document the applications of fragmentation to many rock fracturing processes in which the size distribution is described by a power law. He also showed that such power laws can arise in the Gilvarry's model if $p_f(\ell)$ is constant. For an idealized fragmentation process in which the fragments of a given generation are all of the same relative size, the size distribution is a power law with an exponent τ that depends on p_f .

On the other hand, as described and discussed in Chapter 6, fracture networks of rock often have fractal properties, at least over certain length scales. Thus, a fragmentation process may be used for generating fractal fracture patterns. To do this, Barnsley (1988) proposed the method of iterated function systems by which a fractal-like pattern is obtained from an initial simple shape called the *initiator* by iteratively applying a set of numerical transformations called the *propagators*. Each iteration creates multiple sets of smaller, transformed images that occupy the place of the previous image. After several generations, the set converges to a pattern that may be a fractal. In essence, the method creates fragments of various sizes and shapes. To generate 2D patterns, the two transformations that are used are given by

$$x_n = a_1 x_{n-1} + a_2 y_{n-1} + a_3 x_{n-1} y_{n-1} + a_4, \quad (8.15)$$

$$y_n = a_5 x_{n-1} + a_6 y_{n-1} + a_7 x_{n-1} y_{n-1} + a_8. \quad (8.16)$$

Here, x_n and y_n are the coordinates of a given point in the n th generation, and the a_i s constants of the transformations. If $a_3 = a_7 = 0$, we have linear transformations that can generate self-affine fractals (see Chapters 5 and 6). If, in addition $a_1 = a_6$ and $a_2 = -a_5$, the transformations generate self-similar fractals. Similar transformations can be used for generating 3D patterns.

To make the resulting fracture patterns look more realistic, Acuna and Yortsos (1995) used the fragmentation probability p_f with the above transformations. Specifically, as the fragmentation of the blocks continues, only a fraction p_f of the newly generated blocks is allowed to further fracture. Figure 8.9 shows a 2D example with $p_f = 0.9$, while Figure 8.10 presents a 3D example that has a fractal dimension, $D_f = 2.78$.

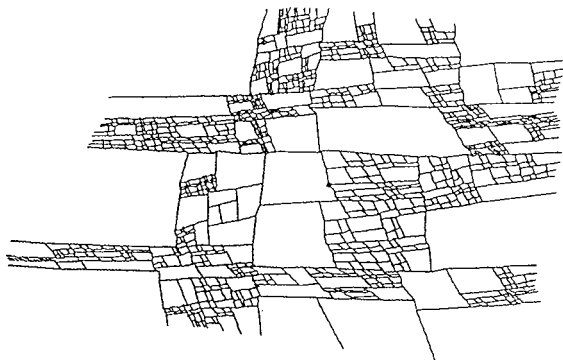


Figure 8.9 A 2D synthetic fractal model of fracture networks with $p_f = 0.9$ (after Acuna and Yortsos, 1995).

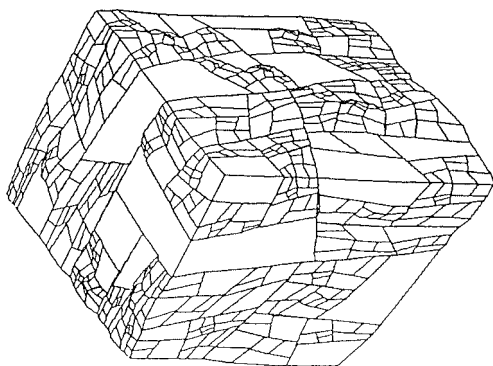


Figure 8.10 A 3D synthetic fractal model of fracture networks (after Acuna and Yortsos, 1995).

Although the method does generate fracture patterns that look realistic and, as discussed by Acuna and Yortsos (1995), some of their hydraulic properties may mimic those of actual fractured rock, it has several shortcomings. There is no clear physical interpretation of the probability p_f , the key parameter for generating fractal fracture patterns and, thus, it is not clear how it can be measured. Moreover, the model contains many adjustable parameters, that is, the a_i s, and it is not clear how they can be estimated if some experimental data for the fracture pattern of a given porous medium, or any other property of the rock are available. In addition, the model completely ignores the physical basis for the fracturing process of porous media, which are the deformation of rock and accumulation of large stresses that result in the nucleation and propagation of the fractures (see below). Finally, the fact that the fractal dimensions of the patterns can be designed to be similar to those of fracture networks of rock does not imply that the flow and transport properties of the patterns are also similar to those of fractured rock. Indeed, many very different patterns may have more or less the same fractal dimension.

A more realistic synthetic model was proposed by Barton *et al.* (1987). In their model, numerical values are selected randomly from the frequency distributions of fracture-trace length, spacing, orientation, crossing, and dead-end fracture intersections, obtained from the analysis of fracture trace maps described in Chapter 6. Such a model represents a more realistic approach to modeling of fracture networks because it incorporates relevant experimental data in the model. Thus, not only will the distributions of various characteristics of the fracture network be automatically in agreement with the data, its fractal dimension for a 2D model will also be in the range, $D_f = 1.3\text{--}1.7$, in agreement with the experimental data described in Chapter 6.

8.5

Mechanical Models of Fracture Networks

The most realistic approach to the modeling of fracture network of porous media is perhaps one that begins with an accurate model of an unfractured porous medium, and then models the process that initiates the microfractures in the porous medium that grow in time, intersect one another, and eventually form a large network. In principle, the fracturing process is highly nonlinear. Moreover, in addition, the fracture process of a sample of natural porous media depends on their depth in the rock as well as their contents, history, and a bewildering number of other parameters. Therefore, generating the fracture network of a given region of porous medium by simulating the complete fracturing process is too difficult and not feasible. It is, however, feasible to carry out such modeling and simulations for a laboratory-scale porous medium. Gertsch (1995) discussed how one may develop 3D fracture network models based on laboratory-scale samples of porous media.

In natural porous media, the presence of the distributed porosity in the form of a large number of flaws with various sizes, shapes, and orientations gives rise to a very complex process of the initiation of a microcrack. Even small initial disorder can be enormously amplified during the fracture propagation, hence implying that fracture of a disordered porous medium (or any solid material for that matter) is a collective phenomenon in which the heterogeneities play a fundamental role. In fact, due to disorder, brittle solids generally exhibit large statistical fluctuations in their fracture strength when nominally identical laboratory samples are tested under identical loading conditions. Because of such statistical fluctuations, it is insufficient, and indeed inappropriate, to represent the fracture behavior of heterogeneous only rock by its *average* properties: fluctuations are important and must not be neglected.

A relatively simple network model was introduced by Sahimi and Goddard (1986) for brittle fracture of disordered solids. In their model the solid is represented by a network in which each bond represents a small portion of the system (for example, the solid matrix of rock). In classical mechanics, a solid is represented by a spring or a beam and, thus, each bond of the network also represents a spring (or a beam). The springs follow the laws of linear elasticity (i.e., force is proportional

to the displacement) up to a critical threshold (defined below), beyond which they break irreversibly and create a microfracture. Heterogeneities may be introduced in the elastic constants and the ultimate strength of the bonds. Thus, the sequence of the bonds that break and the spatial pattern that they form model the regional fracture process. Various properties of such fracture phenomena have been investigated (see, for example, Sahimi and Goddard, 1986; de Arcangelis *et al.*, 1989; Arbabi and Sahimi, 1990b; Sahimi and Arbabi, 1992, 1993, 1996; Sahimi, 1992b; for a comprehensive review, see Sahimi, 2003b). A main advantage of the model is that *any* kind of disorder may be incorporated in the model and its effect on the fracturing process can be evaluated.

To see how the model generates a network of fractures, consider a 2D or 3D network of linear size is L , where L is the number of sites in a given direction. Every site of the network is characterized by a displacement vector $\mathbf{u}_i = (u_{ix}, u_{iy})$, or by $\mathbf{u}_i = (u_{ix}, u_{iy}, u_{iz})$. Here, we consider the case of brittle fracture for which a linear approximation is valid up to a threshold for breaking the springs (defined below). The displacement $\mathbf{u}_i$ is computed by minimizing the elastic energy of the network with respect to $\mathbf{u}_i$, which is given by

$$E = \frac{1}{2} \alpha \sum_{\langle ij \rangle} [(\mathbf{u}_i - \mathbf{u}_j) \cdot \mathbf{R}_{ij}]^2 e_{ij} + \frac{1}{2} \beta \sum_{\langle jik \rangle} (\delta \theta_{jik})^2 e_{ij} e_{ik} . \quad (8.17)$$

The first term of the right side of Eq. (8.17) represents the contribution of stretching or central forces (Hooke's law), whereas the second term is due to the forces that change the angle between two bonds that have a site i in common. Here, α and β are the central and angle-changing force constants, respectively, $\mathbf{R}_{ij}$ a unit vector from site i to site j , e_{ij} the elastic constant of the bond (spring) between i and j , and $\langle jik \rangle$ indicates that the sum is over all triplets in which the bonds ji and ik form an angle with its vertex being at i . Equation (8.17) may be derived by discretizing a continuum equation of elasticity and has rigorous theoretical foundation (Arbabi and Sahimi, 1990a). $\delta \theta_{jik}$ is given by

$$\delta \theta_{jik} = \begin{cases} \frac{(\mathbf{u}_{ij} \times \mathbf{R}_{ij} - \mathbf{u}_{ik} \times \mathbf{R}_{ik}) \cdot (\mathbf{R}_{ij} \times \mathbf{R}_{ik})}{|\mathbf{R}_{ij} \times \mathbf{R}_{ik}|} & \mathbf{R}_{ij} \text{ not } \parallel \text{ to } \mathbf{R}_{ik}, \\ |(\mathbf{u}_{ij} + \mathbf{u}_{ik}) \times \mathbf{R}_{ij}| & \mathbf{R}_{ij} \parallel \text{ to } \mathbf{R}_{ik}, \end{cases} \quad (8.18)$$

where $\mathbf{u}_{ij} = \mathbf{u}_i - \mathbf{u}_j$. For all 2D networks, Eq. (8.18) is simplified to

$$\delta \theta_{jik} = (\mathbf{u}_i - \mathbf{u}_j) \times \mathbf{R}_{ij} - (\mathbf{u}_i - \mathbf{u}_k) \times \mathbf{R}_{ik} . \quad (8.19)$$

Physically, αe_{ij} represents the elastic constant of the bond ij , and $\beta e_{ij} e_{ik}$ the angle-changing force constant of the pair ij and ik . More generally, one may consider a network in which the bonds represent beams that can bend. In that case, one must add the contribution of the torsional or beam-bending forces to Eq. (8.17) (see Sahimi, 2003b, for a comprehensive discussion). Fracture in a network of beams was studied by de Arcangelis *et al.* (1989). It was, however, shown (Arbabi and Sahimi, 1990b) that many properties of fracture processes in the network models, such

as those described below, are independent of whether or not such torsional forces are included in the elastic energy of the system, that is, whether the bonds of the network represent springs or beams. In general, there are at least four ways of incorporating rock heterogeneities in the model.

1. Deletion or suppression of a fraction of the bonds, either at random or in a prescribed manner. The removed bonds represent the pores *before* the fracture process begins.
2. Random or correlated distribution of the elastic constants e_{ij} of the bonds. The idea is that in porous media, the shapes, sizes, and strengths of the regions through which stress is transmitted are broadly distributed, resulting in a different elastic constant e_{ij} for each microscopic region (bond).
3. Random or correlated distribution of the critical thresholds. For example, each bond may be characterized by a critical length ℓ_c such that if it is stretched beyond ℓ_c , it breaks irreversibly. The idea is that even if a porous medium is made up intrinsically of the same material (the same e_{ij} everywhere), it still contains regions that offer different resistances to breakage under an imposed external stress or strain due to, for example, defects or pre-cracked porosity in its structure.
4. The cases 1–3 generate small scale heterogeneities. One way of generating large-scale heterogeneities is by dividing the network into large blocks of many bonds within which the bonds may have the same elastic and fracture threshold (ℓ_c) properties, but between which ℓ_c , e_{ij} , or the porosity (or all of them) varies spatially. The properties of the blocks are correlated with each other. In fact, the analysis of extensive field data for eight oil and gas reservoirs (Sahimi and Tăjer, 2005) indicated that the elastic moduli in large-scale porous media are spatially distributed and follow self-affine fractal distributions of the type described in Chapters 5 and 6. Thus, another way of simulating fracture propagation in natural porous media with large-scale heterogeneities is by distributing the elastic constant e_{ij} of the bonds according to a self-affine distribution. In other words, each bond represents a portion of a porous medium at the length scale for which the field data are measured.

In the present discussion, we consider the case in which a threshold ℓ_c is attributed to each bond which is selected from a given probability density function. While any distribution function may be used, in our discussions, we assume the following

$$f(\ell_c) = (1 - c)\ell_c^{-c}, \quad (8.20)$$

where $c = 0$ corresponds to a uniform distribution in $(0, 1)$. The advantage of this distribution is that by varying c , one can obtain very narrow ($c \simeq 0$) or very broad ($c \simeq 1$) distributions. The same type of distribution, or any other one, may be used for the elastic constants e_{ij} of the bonds. To initiate the fracturing process, a fixed external strain (or stress) is applied to the network in a given direction. To calculate the nodal displacements $\mathbf{u}_i$, the total elastic energy E is minimized with respect to $\mathbf{u}_i$, that is, the equation, $\partial E / \partial \mathbf{u}_i = 0$, is invoked for every node i that, after

writing it down for every interior site of the network, results in a set of about dN simultaneous linear equations for the nodal displacements $\mathbf{u}_i$ of a d -dimensional network of N internal nodes. This set is solved by an efficient numerical method such as the adaptive accelerated Jacobi-conjugate gradient method that uses an acceleration parameter that is optimized for each iteration.

After computing the nodal displacements, the fracture process is initiated by breaking one or more bonds according to a given criterion. If the elastic constants of the bonds are distributed, one can simply break the most stretched bond as it represents the weakest part of the network and is, therefore, the most probable bond to break first. If the critical thresholds ℓ_c are distributed, one may break the bond for which $\ell_c - \ell_s$ is maximum, where ℓ_s is the bond's current length in the deformed network. Doing so would be equivalent to assuming that the rate at which the elastic forces relax throughout the network is much faster than the breaking of one bond. One may also break *all* the bonds with their current lengths exceeding their critical length ℓ_c . Each broken bond represents a microcrack in the network. An advantage of this type of model is that *any* criterion for the nucleation and propagation of the cracks may be used. Moreover, unlike the traditional models of fracture, one does *not* have to begin the simulation with a network in which a microcrack has been inserted in order to ensure the nucleation and propagation of the cracks.

However, while such breaking criteria may be appropriate for a network under shear or tensile forces, they cannot be used for one that is under compression. If one is to simulate such a deformation process, one must use a network of beams because they can also bend and break under compression. In that case, if the bending forces exerted on any beam in the network exceed a critical value, that beam is broken. Alternatively, one may break the beam that suffers the largest bending force. As mentioned, however, the characteristics of fracture networks that are of interest to us in this section and, in particular, the global connectivity of the fracture network, are largely independent of such details.

After a bond (or a beam) is broken, the nodal displacements $\mathbf{u}_i$ are computed again in the new configuration of the network, the next bond to break is identified and broken, the nodal displacements are recomputed, and so on. If the external stress or strain is not large enough to break any new spring (or beam), it is increased gradually. The simulation and the fracturing continue until a sample-spanning network of broken bonds – the fracture network – has been formed.

The most important feature of the model that one is interested in is the structure of the fracture network that results from the model. Moreover, one would like to learn whether the fracture patterns in laboratory-scale porous media differ from those of field-scale porous media. It has been shown that if the parameter c in the distribution (8.20) is small (i.e., a narrow distribution resulting in a weakly disordered model porous medium), a single crack is formed that spans the network with a few smaller cracks that are not necessarily connected to the main sample-spanning fracture. An example is shown in Figure 8.11 that was generated with $c = 0.1$. As c increases towards unity, however, many more cracks, in addition to the

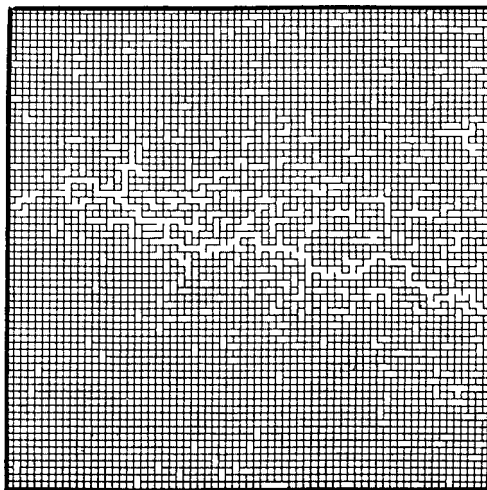


Figure 8.11 A 2D fracture network generated by the mechanical model with $c = 0.1$. The broken bonds represent microfractures.

main sample-spanning fracture, are generated. An example is shown in Figure 8.12 that was generated with $c = 0.9$.

The reason for the differences between the fracture patterns shown in Figures 8.11 and 8.12 is as follows. If c is small, the distribution of the thresholds is narrow, and the network is more or less homogeneous. As soon as the first microfracture is nucleated, the stress enhancement at its tip is larger than at any other part of the network. As a result, it is much more likely that the next microfracture is formed at the tip of the existing fracture. Therefore, a single sample-spanning

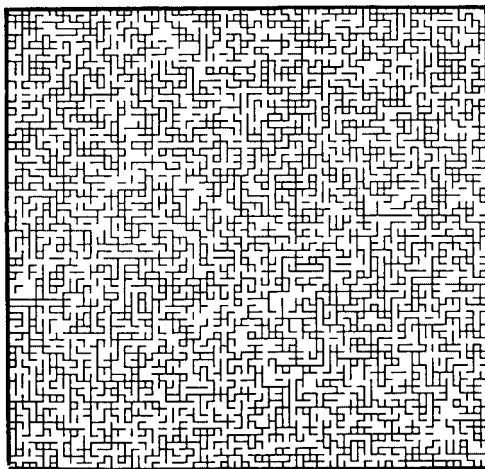


Figure 8.12 A 2D fracture network generated by the mechanical model with $c = 0.9$.

macrofracture is formed (which is not necessarily straight), with the possible exception of a few other microcracks distributed throughout the network. When c is large, however, the distribution of the bonds' strength is very broad, implying that there are many weak as well as strong regions in the network (i.e., the porous medium is highly heterogeneous). Thus, although stress enhancement at the tip of the growing fracture is large, fracture growth may also take place at other regions of the network that are very weak. Thus, formation of the fracture network is more gradual.

The fracture patterns that are formed based on the model that we described are fractal objects, but their fractal dimension D_f depends on the way the patterns are analyzed, and on the spatial distribution of the heterogeneities. One generally finds two distinct fractal dimensions.

1. If one *only* analyzes the fractal scaling of the *single* macrofracture that spans the 2D network, one obtains a fractal dimension $D_f \simeq 1.2 \pm 0.1$, consistent with the data for the surface of a *single major* fault or fracture described in Chapter 5.
2. If, however, one analyzes the statistics of *all* the fractures (broken bonds) of the network (whether or not they are all connected to each other), one finds $D_f \simeq 1.7 \pm 0.05$.

These results are obtained without any large-scale (block) heterogeneities. However, what happens if such heterogeneities are included in the model? To answer this question, we must re-examine the distribution of rock heterogeneities. At small length scales, nucleation and propagation of fractures do not take place at random, but depend on the stress field in the rock because there are not regions with large differences in their mechanical strengths. Once a fracture nucleates in a given block of the rock, stress enhancement at its tip is larger than at any other point of the medium and, therefore, the next microcrack almost surely develops at its tip. As a result, the fractal dimension of the sample-spanning crack is very low, $D_f \simeq 1.2$, because the fracture is almost a straight line.

When there are large-scale (block scale) heterogeneities in the model (porous medium), fracture growth continues within a block or a few blocks until a much stronger region (block) is encountered. When this happens, fracture growth stops and another microfracture nucleates in a weaker region (block) of the porous medium. At the same time, the cracks begin to interact with, and eventually intersect each other. The growth of the new crack also stops when it encounters another strong region (block), and so on. This effect is accentuated if there are correlations between various regions that, as described and discussed in Chapters 5 and 6, is in fact the case.

Thus, it was proposed (Chelidze, 1982; Madden, 1983; Guéguen *et al.*, 1991; Sahimi *et al.*, 1993; Robertson *et al.*, 1995; Renshaw, 1998) that, viewed from large length scales, nucleation and propagation of fractures take place at essentially randomly selected regions of the porous medium because they are more strongly dependent on the spatial distribution of the heterogeneities than on the fluctuations in the stress field. Therefore, the fractal dimension of the fracture network at the largest

length scales should be close to that of the percolation cluster at the percolation threshold because, as described in Chapter 3, in percolation, the bonds of the networks are cut at random, that is, fractures are created randomly.

Others (see, for example, Barton, 1995b) argued that the fracture networks are well connected and, therefore, are well above their percolation threshold. However, their argument may be refuted by, once again, appealing to the physics of fracture propagation. As discussed above, the stress build-up is the driving force for the growth of the cracks. Nevertheless, when a sample-spanning cluster of the connected cracks or fractures is formed for the first time, the built-up stress is relieved and, therefore, there remains little driving force for the cracks to continue growing. Hence, the topology or connectivity of the fracture network should be similar to that of a percolation network at, or at least very near, the percolation threshold. If the growth of the fracture network continues beyond this point, it will likely not be due to mechanical effects.

To make the argument more concrete, we appeal to what is usually referred to as the *critical path analysis* (CPA) that was described in Chapter 6. As described there, it was argued by Ambegaokar *et al.* (1971) and confirmed by many sets of simulations that transport processes, and in particular conduction, in a highly heterogeneous material can be reduced to one in a percolation system at or very near the percolation threshold. The idea is that in a broadly-disordered material, a finite volume fraction of the material possesses a very small conductivity and, hence, makes a negligible contribution to the overall conductivity or other effective flow or transport properties. Therefore, zones of low conductivity may be eliminated from the material which would then reduce it to a percolation system. Ambegaokar *et al.* (1971) described a procedure by which the equivalent percolation network is built up and showed that the resulting percolation system is at, or very near to its percolation threshold. The idea has been applied to fluid flow in laboratory-scale (Katz and Thompson, 1986, 1987; Sahimi, 1993a; Friedman and Seaton, 1998) as well as field-scale porous media (Sahimi and Mukhopadhyay, 1996). We will describe such applications in Chapter 10.

When applied to heterogeneous fractured rock, the CPA indicates that the fracture network should have the morphology of a percolation cluster because the fractures are the main pathways for fluid flow in a fractured rock as their permeabilities (or hydraulic conductances) are much larger than those of the matrix in which they are embedded and, thus, fractured rock may be envisioned as a mixture of nearly impermeable bonds (the pores of the matrix) and the highly permeable bonds (the fractures), that is, a percolation network. Using the procedure of Ambegaokar *et al.* (1971), one then finds that the fracture network of rock must be at, or very near, its percolation threshold.

There is considerable indirect evidence that supports such arguments. For example, as described earlier in this chapter, Long and Billaux (1987) found that at the Fanay-Augères site in France, only about 0.1% of the fractures contributed to fluid flow at large length scales, hence indicating the existence of a barely-connected fracture network which should then be similar to a percolation network at, or very near to, its percolation threshold. Note also that the aforementioned reconstruc-

tion model of Long *et al.* (1991) for identifying the fracture network of rock also indicates that the optimal configuration of the network is similar to a percolation network at or near the percolation threshold, in agreement with the mechanical model of fracture network in the limit of very broad heterogeneities.

8.6

Percolation Properties of Fractures

Regardless of how a fracture network is generated and whether it has to do with mechanical or diagenetic processes, or a combination of both, there is no question that the large-scale connectivity and, hence, the percolation properties of the network is of utmost importance. Thus, in this section, we will describe the percolation properties of a single fracture and a network of fractures computed using various models. When possible, the results are compared with the experimental data.

8.6.1

A Single Fracture

Because the surface of a natural fracture is rough and mostly self-affine, the area of contact and the number of contact areas between the “upper” and “lower” surfaces of a fracture must be statistically-distributed quantities. Two important properties must, therefore, be defined. One is the number of contact zones n_c for an area ℓ^2 ,

$$n_c = N_c \left(\frac{\ell^2}{A} \right), \quad (8.21)$$

where A is the cross-sectional area of the fracture plane, and N_c is the total number of contact over A . The second quantity is the fractional contact area S_c , defined as

$$S_c = \frac{A_c}{A} = 1 - S_0. \quad (8.22)$$

Here, A_0 is the area of the projection of the total contact surface on a reference plane, and $S_0 = A_0/A$ is the fractional void area. In a similar way, the total number N_0 and the number n_0 in an area ℓ^2 of *non-conducting* or *void* zones may be defined as

$$n_0 = N_0 \left(\frac{\ell^2}{A} \right). \quad (8.23)$$

If the two rough surfaces of a fracture are separated, the void zones form a percolation cluster with area A_p in which a fluid can flow. The contact surface may be subdivided into many zones N_c . The percolation cluster of the void zones may be mapped onto a network, which explains why percolation is relevant to describing flow and transport in even a single fracture. If the network has N_b edges (bonds)

and N_s sites (vertices), then the first Betti number $\beta_1(A_p)$ (see Chapter 4) is given by

$$\beta_1(A_p) = N_b - N_s + 1, \quad (8.24)$$

assuming that the number of connected components is one. Recall that the first Betti number is the number of holes through a structure, or the maximum number of non-intersecting closed curves that can be drawn on the surface of the structure without separating it. Equation (8.24) may be further generalized by considering the *whole* void zone of a fracture which consists of some connected components and can be mapped onto a network of edges and sites. The number of such components is the zeroth Betti number β_0 (see Chapter 4), which is related to the first Betti number by

$$\beta_0(A_0) = \beta_1(A_0) - (N_e + N_s). \quad (8.25)$$

Note, however, that the first Betti number β_1 in Eq. (8.25) is defined for the area A_0 .

Now, suppose that w is the *local* separation distance between the upper and lower surfaces of a single fracture, and w_m is its mean value. All the results for a single fracture depend on two parameters, namely, L/l and r/l , where L is the linear size of the sample, r is the resolution, and l is the upper cutoff length for the self-affinity of a fracture's rough surface. Mourzenko *et al.* (1996) showed that

$$n_c \propto \left(\frac{l}{r}\right)^s, \quad (8.26)$$

where the exponent s depends on the Hurst exponent H (see Chapters 5 and 6) that characterizes the self-affinity of the rough surface, and on w_m/σ_h , where σ_h is the standard deviation of the distribution of the surface height h . Mourzenko *et al.* (1996) showed that

$$s = 2(1 - H), \quad (8.27)$$

so that when the surface is no longer self-affine, that is, when $H = 1$, n_c is a constant. Mourzenko *et al.* (1996) also showed that for the percolation cluster in a single fracture,

$$\langle \beta_1(A_p) \rangle \left(\frac{l^2}{A}\right) \sim \left(\frac{l}{r}\right)^{-s_p}, \quad (8.28)$$

where, similar to s , the exponent s_p also depends on H and w_m/σ_h . Power laws (8.26) and (8.28) are similar to those that govern the percolation properties (see Chapter 3), except that the two exponents are nonuniversal, whereas many of the percolation exponents are universal.

Next, one must estimate the percolation threshold of the network for a single fracture. We assume that the connectivity of 3D fracture void space is equivalent

to that of its projected surface on the mean fracture plane. Thus, the percolation of a fracture void (or volume) may be considered as a site (or bond) percolation. Consider the site percolation problem. The probability $p(\mathbf{r})$ that a site at $\mathbf{r}$ is open (occupied) is defined by

$$p(\mathbf{r}) = \begin{cases} 1 & w(\mathbf{r}) > 0, \\ 0 & w(\mathbf{r}) \leq 0. \end{cases} \quad (8.29)$$

Then, the fractional void area S_0 is simply

$$S_0 = \frac{1}{A} \int_A p(\mathbf{r}) dA. \quad (8.30)$$

If the function $w(\mathbf{r})$ is random, then the problem is *exactly* equivalent to a random site percolation described in Chapter 3. If, however, $w(\mathbf{r})$ contains correlations, and in particular, long-range correlations that always exist when the rough surface of a fracture is self-affine, then the problem is equivalent to a site percolation with long-range correlations that was studied by Prakash *et al.* (1992). In this case, the percolation threshold depends on the extent and nature (value of the Hurst exponent H) of the spatial correlations within the sample; see the extensive discussions of such a percolation problem given by Sahimi (1998, 2003a).

Coniglio *et al.* (1977) and Weinrib (1982) showed that the site percolation threshold of continuum systems is exactly $1/2$, a result that was also mentioned in Chapter 3. Thus, the two limits $r/l \gg 1$ and $r/l \ll 1$ (r is the resolution of the discretization of the network of voids) correspond, respectively, to random site percolation ($p_c \simeq 0.593$; see Chapter 3) and continuum systems with $p_c = 1/2$. Sahimi (1994b) and Mourzenko *et al.* (1996) showed that p_c decreases with H when $0.5 < H < 1$, and conversely when $0 < H < 0.5$. Further investigations of the dependence of the percolation threshold p_c in correlated fields were carried out by Schmittbuhl *et al.* (1993b), Sahimi and Mukhopadhyay (1996), Mourzenko *et al.* (1996, 1999), and Marrink *et al.* (2000).

On the other hand, the percolation thresholds $p_c = 1/2$ and 0.593 correspond, respectively, to the mean contact area $S_c = 0.5$ and 0.41 . Do these predictions of percolation agree with the data? Bandis *et al.* (1983) measured S_c for various rock fracture under a normal load and found that it varies between 0.4 and 0.7 , depending on the normal load applied to the sample, in qualitative agreement with the prediction of percolation. In general, the percolation threshold for closing a fracture and preventing fluid flow is reached only at very high loads.

8.6.2

Fracture Networks

In Chapter 3, we described and discussed continuum percolation (for a recent review see Balberg, 2009). Fracture networks represent real-world examples of continuum percolation (Berkowitz, 1995, 2002). In Section 8.2.1, we described the con-

Table 8.1 Percolation thresholds ρ'_c for some 2D and 3D networks of objects. All of the estimates were obtained by numerical simulations (adopted from Adler and Thovert, 1999).

Shape of the object	Spatial dimension	ρ'_c
Orthogonal sticks	2	3.2
Randomly-oriented sticks	2	3.6
Disks or parallel objects	2	4.5
Orthogonal elongated rods	3	0.7
Randomly-oriented elongated rods	3	1.4
Orthogonal squares	3	2
Randomly-oriented squares	2	2.46
Spheres or parallel objects	3	2.80

cepts of excluded volumes and area, which are quite useful for describing the percolation properties of fracture networks. Associated with the excluded volume (or area) is a dimensionless density ρ' (Adler and Thovert, 1999), defined by

$$\rho' = \rho \mathcal{V}_e, \quad (8.31)$$

which represents the number of objects per volume $\mathcal{V}_e$, where ρ is the volumetric density of objects per unit volume. However, the very definition of $\mathcal{V}_e$ also implies that ρ' is also the average number of intersections per object or fracture. Therefore, once we specify the shape of the objects and their orientation distribution – which specify $\mathcal{V}_e$ – Eq. (8.31) would contain information about both volumetric and connectivity (topological) information. For this reason, the determination of the critical value ρ'_c of ρ' – the percolation threshold of the system – as a main continuum percolation property of model fracture networks has been studied quite intensively.

The earliest determination of the percolation thresholds of continuum systems goes back, as mentioned in Chapter 3, to Scher and Zallen (1970), and then later by Pike and Seager (1974) and Haan and Zwanzig (1977). Since then, the percolation thresholds of various continuum systems have been estimated by several research groups and are listed in Table 8.1, as compiled by Adler and Thovert (1999).

For regular polygons with 3–20 vertices as well as for rectangles with an aspect ratio of two, Huseby *et al.* (1997) obtained a nearly constant percolation threshold:

$$\rho'_c \simeq 2.26 \pm 0.04. \quad (8.32)$$

Note that

$$\mathcal{V}_e = \begin{cases} R^3 N_s^2 \cos\left(\frac{\pi}{N_s}\right) \sin^2\left(\frac{\pi}{N_s}\right) & \text{regular } N_s\text{-polygons,} \\ \frac{8R^3 a(a+1)}{(a^2+1)^{\frac{3}{2}}} & \text{rectangles with aspect ratio } a, \end{cases} \quad (8.33)$$

where R is the radius of the fracture (the radius of the circles in which the polygons are embedded), and N_s is the number of the sites or vertices of the polygons. If the rectangles do not have the same size, R^3 is replaced by $\langle R \rangle \langle R^2 \rangle$.

The percolation properties of polydisperse fracture networks with convex polygons distributed randomly and isotropically were studied by Mourzenko *et al.* (2005). First, a volumetric number density of fracture per fracture size, $F(R)$, was defined,

$$F(R) = \rho n(R) . \quad (8.34)$$

Mourzenko *et al.* (2005) studied the case with

$$n(R) = \alpha R^{-\alpha} . \quad (8.35)$$

The polygons were assumed to have identical shapes, but different sizes. Then, an excluded volume was defined by

$$\mathcal{V}_e = \frac{1}{2} v_e (R_1 R_2^2 + R_1^2 R_2) . \quad (8.36)$$

Here, v_e is a dimensionless shape factor with $v_e = \pi^2$, $9\sqrt{3}/2$, and $4\sqrt{2}$ for, respectively, disks, hexagons, and squares. It turns out that for polydisperse fracture networks, the most relevant critical density is given by

$$\rho'_c = \rho_c v_e \langle R^3 \rangle , \quad (8.37)$$

which is different from Eq. (8.31). A cubic domain of linear size L was considered together with $L' = L/R_M$, where R_M is the maximum radius of the fractures. Then, a considerable amount of numerical data were generated using large-scale simulations for several values of L' , the parameter α of the distribution (8.35), and various polygon shapes, including hexagons, triangles, and squares. For example, all the results for $L' = 4$ fell in a narrow range,

$$\rho'_c \simeq 2.95 \pm 0.12 . \quad (8.38)$$

Note that ρ'_c is defined by Eq. (8.37), not Eq. (8.31).

The percolation properties of isotropically-oriented circular cylinders of various aspect ratios were studied by Neda *et al.* (1999) and Sangare and Adler (2009), which are used as a model of thick fractures. In addition, as described in Chapter 6, the lengths of the fractures typically follow a power law and, hence, the percolation threshold should depend on this distribution (Englman *et al.*, 1983; Robinson, 1983, 1984a,b; Charlaix *et al.*, 1984, 1987a; Balberg, 1986; Balberg *et al.*, 1991; Huseby *et al.*, 1997) since even a single long fracture can help a fracture network to become sample-spanning. In particular, Berkowitz (1995) and Watanabe and Takahashi (1995), who studied 2D networks, found the percolation threshold to depend on the second moment of the length distribution.

An alternative general relation for the fracture connectivity was proposed by Berkowitz *et al.* (2000). They suggested that in order to include the effect of the length scale of the measurements, that is, the resolution and the distribution of the

fracture lengths, one should write $n_l(\ell, L)$, the number density of fractures having a length in $[\ell, \ell + d\ell]$, as

$$n_l(\ell, L) = AL^{D_f} \ell^{-a}, \quad (8.39)$$

where D_f is the fractal dimension of the set of intersecting fractures (see Chapter 5). The length scale L (the linear size of the sample) is included here because, as described in Chapter 5, large-scale fracture networks often exhibit fractal behavior. Berkowitz *et al.* (2000) defined the percolation threshold in the traditional way described in Chapter 3, namely, the probability p_c for the formation of a sample-spanning network (which essentially represents a volume fraction). Thus, p_c is defined as

$$p_c = \frac{N_f h^3}{L^3}, \quad (8.40)$$

where N_f is the number of fractures at the percolation threshold, h is the half-width of the fractures, and L is the sample's linear size. Berkowitz *et al.* (2000) then identified p_c as the point at which the fracture network has a connection probability across the sample of $1/2$. For 3D random fractures in the form of ellipses of average accentricity $\langle e \rangle$, de Dreuzay *et al.* (2000) suggested a slightly different expression,

$$p_c = \pi^3 N_f \langle e \rangle \frac{\langle l^3 \rangle}{L^3}, \quad (8.41)$$

where $\langle l^3 \rangle$ is the third moment of the distribution of the fractures' length l .

Complications arise when the length of the fractures is statistically distributed. If the fractures' lengths are all fixed and much smaller than the linear size L of the network, then the standard definition of p_c , Eq. (8.40), is applicable. However, when the fractures can be as long as the system itself (and, therefore, a single fracture can help the fracture network to span), then p_c would be the sum of the two terms that describe the contributions of small and large fractures (Bour and Davy, 1997, 1998). It is also assumed that the probability that the small-scale domains are connected by the presence of the large fractures is small. Hence, $p(L)$, the fracture connectivity, is written as

$$p(L) = \int_{\ell_{\min}}^L \frac{n_l(\ell, L) \ell^{D_f}}{L^{D_f}} d\ell + \int_L^{\ell_{\max}} n_l(\ell, L) d\ell, \quad (8.42)$$

where L^{D_f} represents the volume of the studied area, and $\ell_{\min}$ and $\ell_{\max}$ are, respectively, the minimum and maximum lengths of the fractures in the domain. Assuming that $\ell_{\max} \ll L$ and $p(L) = p_c$, and using Eq. (8.39) for $n_l(\ell, L)$, Eq. (8.42) is solved for the critical length L_c :

$$L_c = \left[(D_f - a + 1)(a - 1) \frac{p_c}{D_f A} + \ell_{\min}^{D_f - a + 1} \frac{(a - 1)}{D_f} \right]^{\frac{1}{D_f - a + 1}}, \quad (8.43)$$

which is valid for $a > 1$ and $a \neq D_f + 1$. Equation (8.43) indicates that if $\ell_{\min}$ is sufficiently small, then, for $1 < a < D_f + 1$, the connectivity threshold is reached at system sizes that depend on the prefactor A (which is related to the density of the fractures). On the other hand, since $L_c > 0$, for $a > D_f + 1$, it follows from Eq. (8.43) that the connectivity threshold is reached when

$$A\ell_{\min}^{D_f-a+1} > (a - D_f - 1)p_c. \quad (8.44)$$

If $a = D_f + 1$, then the fracture network is self-similar in the sense that the number of fractures having lengths that are of the order of the system size is constant. Hence, Eq. (8.42) yields

$$\ln\left(\frac{L}{\ell_{\min}}\right) \geq \frac{p_c}{A} - D_f^{-1}. \quad (8.45)$$

Since $L > \ell_{\min}$, inequality (8.45) is satisfied if $A > p_c D_f$. If, however, $A < p_c D_f$, then the connectivity is reached only for very large systems:

$$L \geq \ell_{\min} \exp\left(\frac{p_c}{D_f} - D_f^{-1}\right). \quad (8.46)$$

Note that inequalities (8.45) and (8.46) indicate that the connectivity of the fracture network is controlled by the *smallest* fractures and, therefore, connectivity can be estimated only by using fracture maps that allow one to observe the smallest fractures. In practice, $\ell_{\min}$ is the resolution length of the map and, therefore, varies with the observation length scale and, hence, with L . In other words, the connectivity depends on $L/\ell_{\min}$.

8.7

Models of Fractured Porous Media

A fracture network alone is used for modeling fluid flow and transport in rock if the matrix has very low permeability. For example, carbonate oil reservoirs of the type that are found in Iran have this property. In this case, the porous matrix acts only as a fluid storage area that feeds the fractures, and is usually assumed to be disconnected. In many practical situations, however, large-scale porous media contain both an extensive fracture network and a porous matrix that does not necessarily have a low permeability. If the matrix can also be sample-spanning, that is, fluid flow through the matrix alone can also occur, it contributes to the overall flow of the fluids in fractured porous medium. This would then constitute the most complex type of porous media, the characterization and realistic modeling of which remain significant challenges, despite the considerable progress that has been made over the last several decades.

Single continuum models – those that represent a fractured porous medium as a single continuum with effective properties – may be utilized under very restrictive conditions. They are valid only when the fracture network is highly dense and

interconnected, or if the interactions between the fractures and the porous matrix allows establishment of local equilibrium. Since one or both of the two conditions are usually not satisfied, more complex continuum models have been developed. Due to the severe shortcomings of the single continuum models, more advanced models began to emerge, beginning perhaps with the early work of (Pollard, 1959) and its extension by Pirson and Pirson (1961).

8.7.1

The Double-Porosity and Double-Permeability Models

The most widely-used models of fractured porous media are the *double-porosity* and *double-permeability* models. The double-porosity model is used when the fracture network is thought to provide the main paths for flow of fluids. Hence, the matrix in this case acts only as a fluid storage area. In the double-permeability model, both the matrix and the fracture network are active in the fluid flow and transport and transport. Some of the earliest papers on more advanced models of fractured porous media are those of Barenblatt *et al.* (1960) and Barenblatt and Zheltov (1960), whose work was extended by Warren and Root (1963) and Odeh (1965).

Barenblatt *et al.* (1960) introduced the double-porosity model. In their model, a fractured rock was envisioned as consisting of two porous systems – the matrix with high porosity and low permeability, and the fractures with low porosity but high permeability. Fluid flow takes place through the two systems separately, and there may be exchange of fluids between the systems at their interface. As such, the model is a continuum one and, similar to any such model, the behavior of a given flow or transport phenomenon is modeled by the classical continuum equations of flow and transport, representing the average behavior of the system, where the averaging is taken with respect to a representative elementary volume (REV).

For fractured porous media, one must have two such REV's, one for the matrix and a second one for the fractures. If one wishes to average the behavior of the system only over one REV, then it must be much larger than the REV used in modeling of flow through unfractured porous media, and it must include many matrix blocks and fractures. Such an approach is usually called *large-scale averaging*, and will be described in Chapter 11 where we consider hydrodynamic dispersion.

Warren and Root (1963) proposed an idealization of the original model of Barenblatt *et al.* In their model, a fractured porous medium is represented by a regular, fully-connected fracture network embedded in a porous matrix represented by parallelepiped blocks, an example of which is shown in Figure 8.13. The matrix is assumed disconnected (not sample spanning), so that fluid flow occurs only through the fracture network. The fracture network and the porous matrix are coupled together by an exchange term, or a *transfer function*, in the governing equations. The precise form of the transfer function is not known. In most applications, it is determined by fitting the governing equations that contain a few adjustable parameters to some available data. It has been claimed that this model is not limited to the idealized structure envisioned by Warren and Root, but in practice, the idealized model has been used most often. A notable feature of the Warren–Root model is

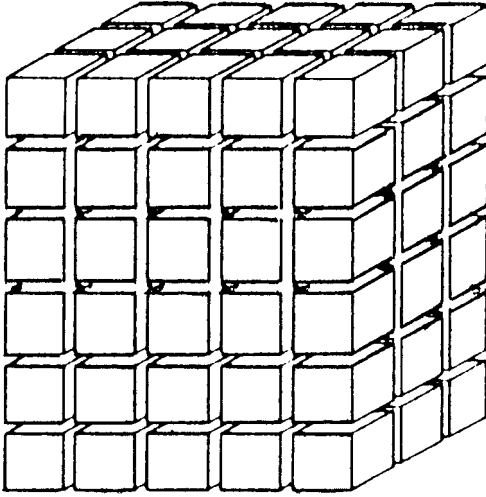


Figure 8.13 The double-porosity model of Warren and Root. The blocks represent the matrix.

that some of its parameters can be estimated from the matrix properties and the size and shape of the blocks. Many variations of such models have been proposed (see, for example, Kazemi, 1969; Kazemi *et al.*, 1969; Crawford *et al.*, 1976; Kazemi *et al.*, 1976; Da Prat *et al.*, 1980; Braester, 1984; Bourbiaux *et al.*, 1998, 1999). In particular, Kazemi (1969) developed a double-porosity model in which the fractures are distributed uniformly throughout the system. Kazemi *et al.* (1969) proposed methods for estimating various parameters of the model.

If the fracture network consists of several parts or zones, each with its own properties, then multicontinuum models have also been proposed (see, for example, Narasimhan and Preuss, 1987; Kazemi and Gilman, 1993). Such fracture networks arise when the fractures have been created by more than one process (for example, diagenetic and mechanical processes). Hughes and Sahimi (1993a,b) proposed a model in which the fracture network, its coupling to the porous matrix, and the connectivity and spatial distribution of the hydraulic conductances in both the fracture network and the matrix are explicitly taken into account. Their model will be described in Chapter 12.

Closmann (1975) and Abdassah and Ershaghi (1986) extended the Warren–Root model by including *three* degrees of porosity. They considered fluid flow in a fractured porous medium in which there exist two kinds of matrix, one having good (high) and a second one poor (low) structural and flow properties. It was assumed that flow between the two matrices was negligible. This model was motivated by investigation of the structure of a fracture network of carbonate rock, and the observations of actual well tests in such reservoirs that indicate anomalous behavior that could not be explained by the double-porosity model of Warren and Root.

If the double-porosity or double-permeability model is to be used in the numerical simulation of fluid flow and transport in large-scale porous media, then the

governing equations would have to be discretized. The most widely-used discretization is the standard finite-difference method, as it gives rise to a sparse and banded matrix that has a simple form. Modern simulators also allow the possibility of using other types of discretization. The effective properties of the blocks are then assigned based on the analysis of the data described in Chapter 5. Thus, from a computational view point, the two models are very efficient. They have, however, several major shortcomings, some of which are as follows.

1. If the fractures are discrete, that is, if they do not form a connected network, the double-porosity models are not valid since these models assume a uniform distribution of fractures everywhere.
2. If fluid flow is dominated by a few connected but very large fractures, then, the double-porosity models are again not useful. As discussed in Section 8.5, the fracture network of rock often has the connectivity of a percolation cluster at, or very close to, the percolation threshold. The active part of the network, that is, its backbone that supports fluid flow and transport processes, contains very few fractures and, therefore, it is natural that a few large-scale fractures dominate the flow of fluids.
3. Although one important ingredient of the double-porosity and double-permeability models is the transfer function that accounts for exchange of fluids between the porous matrix and the fractures, it is not clear in many cases what transfer function one should use.

8.7.2

Discrete Models of Fractured Porous Media

Due to the above shortcomings, models that are based on discrete fracture networks, which were described in Section 8.5, have begun to be used in the numerical simulations of flow and transport in large-scale porous media, such as oil reservoirs and groundwater aquifers. Such models allow one to include the important effect of the structure of fracture network, and in particular, the interconnectivity of the fractures in the simulations. They can be used in several different ways. For example, given enough computational resources, the discrete fracture network models can be used as a complete simulator. Alternatively, they can be used for accurate evaluation of the transfer functions for use in the double-porosity models in order to make such models more accurate and reliable.

Another important aspect of simulation of fluid flow and transport in fractured porous media is the method that is used for discretizing the governing equations. Both regular and irregular discretizations have been used. For example, Lee *et al.* (2001) developed a hierarchical approach in which the small fractures are represented by computed effective properties based on small-scale computations, while large-scale fractures are explicitly included in the model. Since the spatial distribution of fractures in a porous medium is not restricted to those along the three main principal directions (as in the double-porosity models), the most accurate way

of obtaining the numerical solution of the governing equations is based on unstructured discretization. Such methods include finite-element and finite-volume methods. The former has been used by several groups (see, for example, Baca *et al.*, 1984; Kim and Deo, 2000; Riviere *et al.*, 2000; Karimi-Fard and Firoozabadi, 2001; Juanes *et al.*, 2002). We should, however, point out that the finite-element methods may give rise to dense matrices that require intensive computations. In addition, they do not ensure local mass conservation.

As mentioned earlier, Koudina *et al.* (1998) constructed explicit 3D fracture networks in which the fractures were represented by convex polygons. They ignored, however, the matrix. A somewhat similar approach was used by Dershowitz *et al.* (2000), who used a discrete fracture network model to compute the effective flow parameters for use in the double-porosity model. Both groups used a finite-volume approach for solving the governing equations. One can also use a method in which the control volume is aligned with the discontinuities of the permeability field. Caillabet *et al.* (2001) and Granet *et al.* (2001) used such a method, although both groups utilized 2D models.

Karimi-Fard *et al.* (2003) presented a model that represents a refinement of the model of Koudina *et al.* (1998) in that it takes into account the effect of the matrix. The model explicitly includes the effect of matrix-fracture interactions as well as those of fracture-fracture and matrix-matrix. An unstructured discretization method is used. Two-dimensional as well as 3D fractures may be included in the model. The 2D fractures are simply regular or irregular polygons. The 3D fractures are represented by convex polyhedra that are formed by 2D convex polygons.

Finally, Sahimi *et al.* (2010) presented an efficient method for generating a realistic model of fractured porous media in which the fracture network is explicitly accounted for, and the discretization of the matrix is carried by an advancing front technique. We postpone the description of that model to Chapter 15.

9

Single-Phase Flow and Transport in Porous Media: The Continuum Approach

Introduction

Chapter 2 introduced the concept of the permeability of a porous medium, which is a measure of how easily a fluid flows through the medium. In this chapter and the next, we study and discuss single-phase flow and transport in disordered porous media. In both chapters, we focus on low Reynolds-number flows for which Darcy's law is applicable, but we also describe the deviations from such flows. In the present chapter, we focus on describing single-phase flow and transport in what we referred to in Chapter 7 as the *continuum models* of porous media. In Chapter 10, we will study the same phenomena based on the pore network models of disordered porous media. We first present a theoretical derivation of the Darcy's law, and then describe and discuss various methods of estimating the effective permeability and diffusivity of porous media.

9.1

Derivation of Darcy's Law: Ensemble Averaging

As already mentioned in Chapter 2, Darcy's law is expressed as

$$\mathbf{V} = -\frac{K_e}{\mu}(\nabla P - \rho \mathbf{g}), \quad (9.1)$$

where $\mathbf{V}$, μ , and ρ are, respectively, the average velocity, viscosity, and density of the fluid, K_e is the effective permeability, P is the pressure, and $\mathbf{g}$ is the gravity vector. The one-dimensional (1D) version of Eq. (9.1) was discovered empirically by Darcy¹⁾ in 1856. It is its extension to 2D and 3D porous media that is, however, of practical importance. Many derivations of Eq. (9.1) have been presented in the past (Bear, 1972; Gray and O'Neil, 1976; Neuman, 1977; Keller, 1980; Tartar, 1980; Larson, 1981; Whitaker, 1986a; Rubinstein and Torquato, 1989). In this section,

- 1) *Henry Philibert Gaspard Darcy* (1803–1858) was a French engineer. In addition to discovering Darcy's law (originally intended for fluid flow through sands), he made important contributions to hydraulics, including the Darcy–Weisbach equation for calculating the head loss due to friction. The unit of the permeability – *darcy* – is named in his honor.

we present a summary of the derivation of Eq. (9.1) presented by Rubinstein and Torquato (1989), which will also set us up for describing the various estimates of the effective permeability of the continuum models of porous media.

The disordered porous medium is a domain $\mathcal{V}(\omega)$ in the 3D space where ω represents one realization of the medium taken from a probability space ω . We partition the space $\mathcal{V}(\omega)$ into two parts: the pore region $\mathcal{V}_1(\omega)$ with porosity (volume fraction) ϕ , and the solid matrix region $\mathcal{V}_2(\omega)$ with volume fraction $1 - \phi$. Suppose that $\partial\mathcal{V}(\omega)$ is the surface between the two regions. The pore space is described by a characteristic function $I(\mathbf{r}, \omega)$, defined by

$$I(\mathbf{r}, \omega) = \begin{cases} 1, & \mathbf{r} \in \mathcal{V}_1(\omega) \\ 0, & \mathbf{r} \in \mathcal{V}_2(\omega) \end{cases} \quad (9.2)$$

At the microscopic scale and under the steady-state condition, the flow of a Newtonian fluid is described by the Stokes' and continuity equations:

$$\mu \nabla^2 \mathbf{v} = \nabla P \quad \text{in } \mathcal{V}_1, \quad (9.3)$$

$$\nabla \cdot \mathbf{v} = 0 \quad \text{in } \mathcal{V}_1, \quad (9.4)$$

$$\mathbf{v} = \mathbf{0} \quad \text{on } \partial\mathcal{V}, \quad (9.5)$$

where $\mathbf{v}$ is the *microscopic* fluid velocity. The macroscopic length scale of the porous medium – on which the applied pressure varies – is L , while the microscopic length scale – over which the characteristic function I changes – is l . Thus, there is a small parameter, $\epsilon = l/L$, associated with rapid fluctuations in the morphology of the pore region $\mathcal{V}_1(\omega)$ (due to porous medium being disordered). Then, the velocity and pressure depend on *two* scales: a slow scale $\mathbf{x}$, and a fast one $\mathbf{y} = \mathbf{x}/\epsilon$. Thus,

$$\mu \nabla^2 \mathbf{v}(\mathbf{x}, \mathbf{y}, \omega) = \nabla P(\mathbf{x}, \mathbf{y}, \omega) \quad \text{in } \mathcal{V}_1^\epsilon(\omega), \quad (9.6)$$

$$\nabla \cdot \mathbf{v}(\mathbf{x}, \mathbf{y}, \omega) = 0 \quad \text{in } \mathcal{V}_1^\epsilon(\omega), \quad (9.7)$$

$$\mathbf{v}(\mathbf{x}, \mathbf{y}, \omega) = \mathbf{0} \quad \text{on } \partial\mathcal{V}. \quad (9.8)$$

To derive the governing equations at the macroscopic scale, a two-scale expansion is utilized (Sanchez-Palencia, 1980):

$$\mathbf{v}(\mathbf{x}, \mathbf{y}, \omega) = \epsilon^2 \mathbf{v}_0(\mathbf{x}, \mathbf{y}, \omega) + \epsilon^3 \mathbf{v}_1(\mathbf{x}, \mathbf{y}, \omega) + \dots, \quad (9.9)$$

$$P(\mathbf{x}, \mathbf{y}, \omega) = P_0(\mathbf{x}) + \epsilon P_1(\mathbf{x}, \mathbf{y}, \omega) + \dots \quad (9.10)$$

The reason that the first term in the expansion for the velocity is $O(\epsilon^2)$ is that the pores are small, and the velocity must obey the no-slip boundary condition on the pores' surface, that is, Eq. (9.5).

If we substitute the expansions for $\mathbf{v}$ and P into the Stokes' and continuity equations, and equate the coefficients of equal powers of ϵ , we obtain

$$\mu \nabla_{\mathbf{y}}^2 \mathbf{v}_0(\mathbf{x}, \mathbf{y}, \omega) = \nabla_{\mathbf{y}} P_1(\mathbf{x}, \mathbf{y}, \omega) + \nabla_{\mathbf{x}} P_0(\mathbf{x}), \quad (9.11)$$

$$\nabla_{\gamma} \cdot \mathbf{v}_0(\mathbf{x}, \gamma, \omega) = 0, \quad (9.12)$$

$$\nabla_{\mathbf{x}} \cdot \mathbf{v}_0(\mathbf{x}, \gamma, \omega) + \nabla_{\gamma} \cdot \mathbf{v}_1(\mathbf{x}, \gamma, \omega) = 0. \quad (9.13)$$

Note that the relation, $\nabla = \nabla_{\mathbf{x}} + \epsilon^{-1} \nabla_{\gamma}$, has been utilized. To obtain the macroscopic form of the governing equations, one must carry out an ensemble averaging, that is, average the above equations over all the possible realizations of the pore space that can be obtained from the probability space ω . To do so requires making an additional assumption, namely, that the porous medium is *locally stationary* (on the γ -scale), that is, its averaged properties do not depend on length scales.

We consider the most general case in which the porous medium may be anisotropic with direction-dependent permeabilities. Thus, subject to the assumption of local stationarity, ensemble averaging of Eq. (9.13) yields

$$\nabla_{\mathbf{x}} \cdot \mathbf{V}(\mathbf{x}) + \langle \nabla_{\gamma} \cdot \mathbf{V}_1(\mathbf{x}, \gamma, \omega) \rangle = 0, \quad (9.14)$$

$$\mathbf{V}(\mathbf{x}) = \langle \mathbf{v}_0 \rangle(\mathbf{x}), \quad (9.15)$$

where $\langle \cdot \rangle$ denotes an ensemble average. It is straightforward to show that the second term of Eq. (9.14) is zero. Thus,

$$\nabla_{\mathbf{x}} \cdot \mathbf{V}(\mathbf{x}) = 0. \quad (9.16)$$

Two stationary random functions, $\mathbf{w}(\gamma, \omega) = [w_{ij}]$, and $\boldsymbol{\pi}(\gamma, \omega) = [\pi_i]$ are now introduced that are the solutions of

$$\nabla_{\gamma}^2 \mathbf{w} = \nabla_{\gamma} \boldsymbol{\pi} - \mathbf{I} \quad \text{in } \mathcal{V}_1(\omega), \quad (9.17)$$

$$\nabla_{\gamma} \cdot \mathbf{w} = 0 \quad \text{in } \mathcal{V}_1(\omega), \quad (9.18)$$

$$\mathbf{w} = \mathbf{0} \quad \text{on } \partial \mathcal{V}(\omega), \quad (9.19)$$

where $\mathbf{I}$ is the unit dyadic. We assume that $\mathbf{w}$ and $\mathbf{P}$ are zero in the matrix phase. Then, it is straightforward to show that the functions $\mathbf{v}_0(\mathbf{x}, \gamma, \omega)$ and $P_1(\mathbf{x}, \gamma, \omega)$ may be written as

$$\mathbf{v}_0(\mathbf{x}, \gamma, \omega) = -\frac{1}{\mu} \nabla P_0(\mathbf{x}) \cdot \mathbf{w}(\gamma, \omega), \quad (9.20)$$

$$P_1(\mathbf{x}, \gamma, \omega) = \nabla P_0(\mathbf{x}) \cdot \boldsymbol{\pi}(\gamma, \omega). \quad (9.21)$$

If we ensemble average Eq. (9.20), we obtain

$$\mathbf{V}(\mathbf{x}) = -\frac{1}{\mu} \langle \mathbf{w}(\gamma, \omega) \rangle \cdot \nabla P_0(\mathbf{x}), \quad (9.22)$$

which is the *macroscopic* Darcy's law and, when compared with Eq. (9.1), implies that the effective permeability tensor is given by

$$\mathbf{K}_e = \langle \mathbf{w}(\gamma, \omega) \rangle. \quad (9.23)$$

If the porous medium is *macroscopically* isotropic, then $\mathbf{K}_e = K_e \mathbf{I}$, with $K_e = 1/3(\mathbf{w} : \mathbf{I})$. Note that the effective permeability tensor $\mathbf{K}_e$ is *symmetric* and *positive definite*.

Such a derivation of Darcy's law allows one to *exactly* calculate the effective permeability $\mathbf{K}_e$ if the boundary-value problem described by Eqs. (9.17)–(9.19) can be solved. In practice, however, this is extremely difficult, if not impossible, unless the microstructure of the porous medium is simple enough. Thus, over the years, many empirical or semi-empirical expressions for the effective permeability of disordered porous media have been proposed that are described by Bear (1972) and Scheidegger (1974). We describe and discuss the more recent results for the effective permeability of the continuum models of disordered porous media.

9.2

Measurement of Permeability

If a porous medium is isotropic, then a cylindrical core sample is usually utilized for measuring its effective permeability. The core may be horizontal or vertical. Both liquids and gases are used for measuring K_e . The disadvantage of using a liquid is that it may cause swelling of the pore structure (that can happen in clays, for example), or carry ions that react with the pore structure. According to Darcy's law, a single steady-state flow rate should be sufficient for measuring the effective permeability, though, in practice, one encounters significant experimental errors. Thus, one must carry out a series of measurements at various pressure gradients, measure the volume flow rate, and from the best straight-line fit of the data estimates K_e . Significant deviations from the linearity indicate that the flow is outside the regime of validity of Darcy's law.

If a gas is used for measuring the effective permeability, then its compressibility must be taken into account. The compressibility affects the measurements significantly, as the volume flow rate and the velocity depend on the pressure and vary from one face of the sample to the opposite face. To address this problem, we use the fact that for flow of a gas at constant temperature and under steady-state condition, the product of the velocity and the pressure is constant. Thus, we obtain

$$V_z^{(2)} = - \left(\frac{K_e}{\mu} \right) \left(\frac{P_2^2 - P_1^2}{2P_2 L} \right),$$

where $V_z^{(2)}$ is the fluid velocity at point two (the exit), and z is the direction of macroscopic flow. Thus, the above equation, which expresses the exit flow velocity in terms of the entrance and exit pressures, may be used for estimating the effective permeability. However, using a gas gives rise to another complication. That is the so-called *Klinkenberg effect*, whereby the gas molecules may not obey the no-slip boundary condition on the pores' walls. The net effect is that the effective permeability may have an apparent dependence on the pressure. In such cases, the

appropriate equation is given by

$$V_z^{(2)} = -\frac{K_e}{\mu} \left(1 + \frac{b}{P_1 + P_2} \right) \left(\frac{P_1^2 - P_2^2}{2P_2 L} \right),$$

where b is a constant characteristic of the gas and the porous medium. Unsteady-state methods may also be used for measuring the effective permeability; see, for example, Haskett *et al.* (1986).

9.3

Exact Results

Before describing the exact results, we should first precisely define what we mean by exact. Aside from some very general results that are not usually useful for quantitative estimation of the effective permeability, there currently exists no exact and *explicit* expression for the effective permeability of disordered porous media with an *arbitrary microstructure*. Therefore, when we refer to exact results, we mean those for a *given* morphology of the pore space. There are also some rigorous upper and lower bounds to the effective permeability that are applicable to disordered porous media with an arbitrary microstructure. However, in most cases, they are useful only for obtaining estimates of K_e for models of porous media with relatively simple microstructures.

Almost all the available exact results for the effective flow and transport properties of continuum models of porous media are for those in which fluid flow or transport occurs in the pore space *between* spatially-periodic arrays of spherical or cylindrical particles of radius R , with their centers placed at the nodes of a regular lattice, for example, the simple-cubic (SC) lattice. We assume that the volume fraction of the particles is $\phi_2 = 1 - \phi$, where ϕ the porosity.

9.3.1

Fluid Flow

Hasimoto (1959) was the first to analyze slow fluid flow through a dilute ($\phi_2 = 1 - \phi \ll 1$) cubic array of spheres. He first derived the periodic fundamental solution to the Stokes' equation, and then expanded the flow velocity in terms of that solution in order to obtain an expression for the effective permeability K_e of the array. Sangani and Acrivos (1982) (see also Zick and Homsy, 1982) modified and extended Hasimoto's work, and derived expressions for the permeability of all the three cubic lattices of spheres, namely, the SC, the body-centered and the face-centered cubic lattice (BCC and FCC, respectively). We describe their approach in detail as it represents a highly versatile and valuable method for analyzing such problems.

As usual, under the steady-state condition, the equation to be solved is the Stokes' equation coupled with the continuity equation, Eqs. (9.3)–(9.5). Thus, if V_i is the

component of the fluid velocity in the x_i -direction, then

$$\mu \nabla^2 V_i = \frac{\partial P}{\partial x_i} , \quad (9.24)$$

$$\frac{\partial V_i}{\partial x_i} = 0 . \quad (9.25)$$

Since the lattice is periodic and symmetric, the velocity components should satisfy the following equations (Sørensen and Stewart, 1974)

$$V_1(x_1, x_2, x_3) = V_1(-x_1, x_2, x_3) = V_1(x_1, -x_2, x_3) = V_1(x_1, x_2, -x_3) , \quad (9.26)$$

$$V_2(x_1, x_2, x_3) = -V_2(-x_1, x_2, x_3) = -V_2(x_1, -x_2, x_3) = -V_2(x_1, x_2, -x_3) , \quad (9.27)$$

$$V_3(x_1, x_2, x_3) = -V_3(-x_1, x_2, x_3) = V_3(x_1, -x_2, x_3) = -V_3(x_1, x_2, -x_3) , \quad (9.28)$$

$$V_2(x_1, x_2, x_3) = V_3(x_1, x_3, x_2) , \quad (9.29)$$

$$V_1(x_1, x_2, x_3) = V_1(x_1, x_3, x_2) , \quad (9.30)$$

$$\mathbf{V}(\mathbf{r} + \mathbf{r}_n) = \mathbf{V}(\mathbf{r}) , \quad (9.31)$$

where $\mathbf{r}_n$ is a vector denoting the center of a sphere. The variables are now made dimensionless: all the distances by a length h (for example, the distance between the centers of two neighboring particles in the lattice), the velocity components with the mean flow velocity V_m , and the pressure with $\mu V_m / h$. Thus, unless specified otherwise, from now on, all the quantities are dimensionless.

The periodic fundamental solution of the problem is obtained by solving

$$\nabla^2 V_i = \frac{\partial P}{\partial x_i} + \delta_{i1} \sum_n \delta(\mathbf{r} - \mathbf{r}_n) , \quad (9.32)$$

$$\frac{\partial V_i}{\partial x_i} = 0 , \quad (9.33)$$

where $\delta(\mathbf{r} - \mathbf{r}_n)$ is the Dirac delta function. Hasimoto (1959) showed that

$$V_i = V_m \delta_{i1} - \frac{1}{4\pi} \left(S_1 \delta_{i1} - \frac{\partial^2 S_2}{\partial x_1 \partial x_i} \right) , \quad (9.34)$$

$$\frac{\partial P}{\partial x_i} = -\frac{\delta_{i1}}{\mathcal{V}_c} + \frac{\partial^2 S_1}{\partial x_1 \partial x_i} , \quad (9.35)$$

where $\mathcal{V}_c$ is the dimensionless volume of the basic unit cell of the lattice with $\mathcal{V}_c = 1, 1/2$, and $1/4$ for the SC, BCC and FCC lattices, respectively. The functions S_1 and S_2 are the solutions of the following equations

$$\nabla^2 S_2 = S_1 , \quad (9.36)$$

$$\nabla^2 S_1 = 4\pi \left[\frac{1}{\mathcal{V}_c} - \sum_n \delta(\mathbf{r} - \mathbf{r}_n) \right] , \quad (9.37)$$

and are given by

$$S_1 = \frac{1}{\pi \mathcal{V}_c} \sum_{\mathbf{k}_n \neq 0} k_n^{-2} \exp[-2\pi i (\mathbf{k}_n \cdot \mathbf{r})] , \quad (9.38)$$

$$S_2 = -\frac{1}{4\pi^3 \mathcal{V}_c} \sum_{\mathbf{k}_n \neq 0} k_n^{-4} \exp[-2\pi i (\mathbf{k}_n \cdot \mathbf{r})] , \quad (9.39)$$

where $i = \sqrt{-1}$. Here, $\mathbf{k}_n$ are vectors in the reciprocal lattices,

$$\mathbf{k}_n = \sum_{i=1}^3 n_i \mathbf{b}_i , \quad (9.40)$$

where $\mathbf{b}_i$ are the unit vectors in the reciprocal lattices,

$$\mathbf{k}_n \cdot \mathbf{a}_i = n_i , \quad i = 1, 2, 3 , \quad (9.41)$$

and $\mathbf{a}_i$ are the unit vectors in the original lattices. For the SC lattice, $\mathbf{a}_1 = \mathbf{b}_1 = (1, 0, 0)$, $\mathbf{a}_2 = \mathbf{b}_2 = (0, 1, 0)$, and $\mathbf{a}_3 = \mathbf{b}_3 = (0, 0, 1)$. For the BCC lattice,

$$\mathbf{a}_1 = \frac{1}{2}(1, 1, -1) , \quad \mathbf{a}_2 = \frac{1}{2}(-1, 1, 1) , \quad \mathbf{a}_3 = \frac{1}{2}(1, -1, 1) , \quad (9.42)$$

$$\mathbf{b}_1 = (1, 1, 0) , \quad \mathbf{b}_2 = (0, 1, 1) , \quad \mathbf{b}_3 = (1, 0, 1) , \quad (9.43)$$

while for the FCC lattice, $(\mathbf{a}_i)_{\text{FCC}} = \frac{1}{2}(\mathbf{b}_i)_{\text{BCC}}$, and $(\mathbf{b}_i)_{\text{FCC}} = 2(\mathbf{a}_i)_{\text{BCC}}$.

To derive the complete solution for $\mathbf{V}$ and P , Sangani and Acrivos (1982) added to the fundamental solution the derivatives of $\mathbf{V}$ and S_1 multiplied by some unknown coefficients. From the sum, the terms that did not satisfy the Stokes' and continuity equations and the symmetry conditions (9.26)–(9.31) were subtracted. Then, the complete solution would have the following form, assuming that the mean flow is in the x_1 direction,

$$V_1 = V_m - \frac{1}{4\pi} \left[G_1 \left(S_1 - \frac{\partial^2 S_2}{\partial x_1^2} \right) + G_2 \frac{\partial^2 S_1}{\partial x_1^2} - G_3 \left(\frac{\partial^4}{\partial x_2^4} - 6 \frac{\partial^4}{\partial x_2^2 \partial x_3^2} + \frac{\partial^4}{\partial x_3^4} \right) S_1 \right] , \quad (9.44)$$

$$V_2 = \frac{1}{4\pi} \left[G_1 \frac{\partial^2 S_2}{\partial x_1 \partial x_2} - G_2 \frac{\partial^2 S_1}{\partial x_1 \partial x_2} - G_3 \frac{\partial}{\partial x_1} \left(\frac{\partial^3}{\partial x_2^3} - 3 \frac{\partial^3}{\partial x_2^2 \partial x_3} \right) S_1 \right] , \quad (9.45)$$

$$V_3 = \frac{1}{4\pi} \left[G_1 \frac{\partial^2 S_2}{\partial x_1 \partial x_3} - G_2 \frac{\partial^2 S_1}{\partial x_1 \partial x_3} - G_3 \frac{\partial}{\partial x_1} \left(\frac{\partial^3}{\partial x_3^3} - 3 \frac{\partial^3}{\partial x_2^2 \partial x_3} \right) S_1 \right] , \quad (9.46)$$

$$\frac{\partial P}{\partial x_i} = -\frac{6\pi R K}{h \mathcal{V}_c K_s} \delta_{i1} + \frac{1}{4\pi} G_1 \frac{\partial^2 S_1}{\partial x_1 \partial x_i} . \quad (9.47)$$

Here, G_1 is an operator given by

$$G_1 = \sum_{M=0}^{\infty} \sum_{m=0}^{m \leq M/2} A_{nm} \left\{ \frac{\partial^{2n}}{\partial x_1^{2n}} \left[\left(\frac{\partial}{\partial \xi_1} \right)^{4m} + \left(\frac{\partial}{\partial \xi_2} \right)^{4m} \right] \right\} , \quad (9.48)$$

with similar expressions for G_2 and G_3 , but with corresponding coefficients B_{nm} and C_{nm} , where $\xi_1 = x_2 + ix_3$, $\xi_2 = x_2 - ix_3$, and $M = n + 2m$. Here, K_s is called the *Stokes' permeability* with $K_s = 2R^2/(9\phi_2)$ (see below). The coefficients A_{nm} , B_{nm} and C_{nm} are unknown and are determined by using the no-slip boundary condition. The effective permeability and the average flow velocity are then given by (Sangani and Acrivos, 1982)

$$K_e = K_s \frac{hA_{00}}{3\pi R}, \quad (9.49)$$

$$V_m = 1 + \frac{2B_{00}}{\mathcal{V}_c}. \quad (9.50)$$

Therefore, knowledge of A_{00} and B_{00} suffices for determining K_e and V_m . The final expression for the permeability of a SC array of spheres is

$$\frac{K_s}{K_e} = 1 - 1.7601\phi_2^{\frac{1}{3}} + \phi_2 - 1.5593\phi_2^2 + 3.9799\phi_2^{\frac{8}{3}} - 3.0734\phi_2^{\frac{10}{3}} + \mathcal{O}\left(\phi_2^{\frac{11}{3}}\right). \quad (9.51)$$

Sangani and Acrivos (1982) also derived the numerical coefficients in the expansion (51) for the BCC and FCC lattices. Their results are convergent for $0 < \phi_2/\phi_{\max} < 0.85$, where $\phi_{\max}$ is the maximum packing fraction with $\phi_{\max} = \pi/6$, $\sqrt{3}\pi/8$, and $\sqrt{2}\pi/6$ for the SC, BCC, and FCC lattices, respectively. They also agree with the experimental data reported by Martin *et al.* (1951) and Susskind and Becker (1967).

The problem of fluid flow in the pore space between a square or hexagonal array of parallel circular (solid) cylinders can be analyzed by the same method. Assume that the mean flow is in the x_1 direction, and that the axes of the cylinders are parallel to the x_3 direction. In this case, $V_3 = 0$, and the general solutions for V_1 and V_2 are given by Eqs. (9.44) and (9.45), provided that we set $G_3 \equiv 0$. Then, the operator G_1 would have the following form

$$G_1 = \sum_{n=0}^{\infty} A_n \frac{\partial^{2n}}{\partial x_1^{2n}}, \quad (9.52)$$

with a similar expression for G_2 , but with corresponding coefficients B_n . The coefficients A_n and B_n are determined by applying the no-slip boundary condition. The final results are (Sangani and Acrivos, 1982)

$$\frac{K_s}{K_e} = -\frac{1}{2} \ln \phi_2 - 0.738 + \phi_2 - 0.887\phi_2^2 + 2.039\phi_2^3 + \mathcal{O}(\phi_2^4), \quad (9.53)$$

$$\frac{K_s}{K_e} = -\frac{1}{2} \ln \phi_2 - 0.745 + \phi_2 - \frac{1}{4}\phi_2^2 + \mathcal{O}(\phi_2^4), \quad (9.54)$$

for the square and hexagonal arrays, respectively, in which ϕ_2 represents the *area fraction* of the particles.

The results derived so far pertain to single-phase Darcy flow through unconsolidated porous media. In an attempt to study the problem in consolidated porous media, Larson and Higdon (1989) analyzed a problem in which the spheres in a cubic packing were allowed to grow. Figure 9.1 shows the model they considered. They expressed the velocity and pressure fields in terms of the harmonic expansions in spherical coordinates, as described by Happel and Brenner (1983). In this method, one writes

$$P = \sum_{n=-\infty}^{\infty} p_n, \quad (9.55)$$

$$\mathbf{V} = \sum_{n=-\infty}^{\infty} [\nabla \times (\mathbf{r}\chi_n) + \nabla\psi_n + \alpha_n r^2 \nabla p_n + \beta_n \mathbf{r} p_n], \quad (9.56)$$

where $\alpha_n = (n+3)/2(n+1)(2n+3)\mu$, $\beta_n = n/(n+1)(2n+3)\mu$, and p_n , χ_n , and ψ_n are solid spherical harmonics which are written in terms of the associated Legendre functions²⁾ P_n^m in the form $r^n P_n^m(\cos\theta) \exp(im\psi)$. Equations (9.55) and (9.56) were then substituted in the continuity and Stokes' equations, the geometry of the packing was specified, and the numerical solution of the flow problem was obtained. At low porosities (high volume fractions of the spheres), an analytical asymptotic expression was also derived by Larson and Higdon.

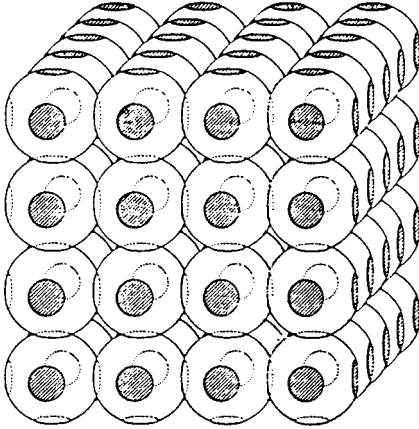


Figure 9.1 Schematic view of the model of consolidated porous media studied by Larson and Higdon.

- 2) Recall that the **Legendre differential equation** for $y = P_n^m$ is given by

$$(1-r^2)y'' - 2ry' + \left[n(n+1) - \frac{m^2}{1-r^2} \right] y = 0.$$

See Dunster (2010) for more details.

9.3.2

Transport

Similar to the fluid flow problem, conduction in the same type of periodic arrays has been studied for a long time. First, consider a SC lattice of identical spheres inserted in a uniform matrix. The ratio of the conductivities of the spheres and the matrix is σ , and we wish to determine the effective conductivity g_e of the composite normalized by that of the matrix. If the spheres have a small conductivity and the matrix has a large one, then σ is small and the results are directly applicable to conduction in fluid-saturated unconsolidated porous media as well as diffusion in the same porous media (by virtue of Einstein's relation that relates the effective conductivity and diffusivity; see Chapter 3). In addition, as described in Section 9.8, there is an approximate but accurate relation between g_e and K_e . Therefore, results for the former may be used for obtaining estimates for the latter.

The spheres' radius is R , measured in units of the lattice spacing and, thus, their volume fraction is $\phi_2 = 4\pi R^3/3$. Due to the symmetry of the lattice, the effective conductivity of the composite is independent of the direction along which an external field E_0 is applied and, therefore, we assume that E_0 is applied in, say, the x direction. The most convenient coordinate system relative to the center of a sphere is the spherical coordinates (r, θ, φ) , where θ is measured from the x axis and φ is an azimuthal angle measured from the xy plane.

Under steady-state condition, the potential (voltage, temperature, etc.) distribution in the material satisfies the Laplace equation, $\nabla^2 E = 0$. To compute g_e , one must determine the potential distribution both inside and outside the spheres. The first investigations of this problem date back to the nineteenth century (see Sahimi, 2003a, and references therein), and have been studied ever since by a large number of research groups. The exact solution of the problem was derived by McPhedran and McKenzie (1978). We do not give the details of their derivation, which are long. The interested reader can consult the original reference as well as Sahimi (2003a). From the exact solution, McPhedran and McKenzie (1978) obtained the following very accurate approximant (derived by truncating the exact solution that was in the form of an infinite series):

$$g_e = 1 - 3 \frac{\phi_2}{D}, \quad (9.57)$$

with

$$D = \mathcal{R}_1^{-1} + \phi_2 - b_1 \mathcal{R}_5 \phi_2^{\frac{14}{3}} - c_1 \mathcal{R}_7 \phi_2^6 - a_1 \phi_2^{\frac{10}{3}} \frac{1 - c_2 \mathcal{R}_5 \phi_2^{\frac{11}{3}} + c_3 \mathcal{R}_5^2 \phi_2^{\frac{22}{3}}}{\frac{1}{\mathcal{R}_3} + b_2 \phi_2^{\frac{7}{3}} - c_4 \mathcal{R}_5 \phi_2^6} \quad (9.58)$$

where,

$$\mathcal{R}_n = \frac{1 - \sigma}{\sigma + 1 + n^{-1}}, \quad (9.59)$$

and $a_1 = 1.305$, $b_1 = 0.01479$, $b_2 = 0.4054$, $c_1 = 0.1259$, $c_2 = 0.5289$, $c_3 = 0.06993$, and $c_4 = 6.1673$.

We should also mention some of the important results that have been derived by other groups. Zuzovsky and Brenner (1977) presented the following expression for the effective conductivity of the SC array of spheres:

$$g_e = 1 - 3\phi_2 \left[\frac{2 + \sigma}{1 - \sigma} + \phi_2 - \frac{16d_1^2 \phi_2^{\frac{10}{3}}}{\frac{1}{\mathcal{R}_3} + 20d_2 \phi_2^{\frac{7}{3}}} - 53.5d_2^2 \mathcal{R}_5 \phi_2^{\frac{14}{3}} + \mathcal{O}(\phi_2^6) \right]^{-1}, \quad (9.60)$$

where $d_1 = 0.2857$ and $d_2 = 0.02036$. The predictions of Eq. (9.60) agree closely with those of Eqs. (9.57) and (9.58). In the limit $\sigma \rightarrow 0$, that is, when the spheres become insulating, Eq. (9.60) reduces to

$$g_e = 1 - 3\phi_2 \left[2 + \phi_2 - \frac{1.306 \phi_2^{\frac{10}{3}}}{\frac{4}{3} + 0.407 \phi_2^{\frac{7}{3}}} - 0.018 \phi_2^{\frac{14}{3}} + \mathcal{O}(\phi_2^6) \right]^{-1}, \quad (9.61)$$

the predictions of which agree with the experimental data of Meredith and Tobias (1960). In the opposite limit of $\sigma \rightarrow \infty$, that is, when the spheres become perfectly conducting, Eq. (9.60) reduces to

$$g_e = 1 + 3\phi_2 \left[1 - \phi_2 - \frac{1.306 \phi_2^{\frac{10}{3}}}{1 - 0.407 \phi_2^{\frac{7}{3}}} - 0.022 \phi_2^{\frac{14}{3}} + \mathcal{O}(\phi_2^6) \right]^{-1}, \quad (9.62)$$

the predictions of which also agree with the experimental data of Meredith and Tobias (1960) for ϕ_2 as large as 0.45.

Sangani and Acrivos (1983) presented expressions similar to Eqs. (9.60)–(9.62), except that theirs were accurate to order $\mathcal{O}(\phi_2^{25/3})$. They determined the thermal conductivity g_e of a regular packing of spheres of conductivity g_2 embedded in a matrix of unit conductivity. Their result is given by

$$g_e = 1 - 3\phi_2 \left[-\frac{1}{L_1} + \phi_2 + c_1 \frac{L_2 \phi_2^{\frac{10}{3}} \left(1 + c_2 L_3 \phi_2^{\frac{11}{3}} \right)}{1 - c_3 L_2 \phi_2^{\frac{7}{3}}} + \sum_{n=4}^6 c_n L_{n-1} \phi_2^{\frac{4n-2}{3}} \right]^{-1}, \quad (9.63)$$

where the higher order terms in the expansion are of the order of $\mathcal{O}(\phi_p^{25/3})$,

$$L_n = \frac{g_2 - 1}{g_2 + \frac{2n}{2n-1}}, \quad (9.64)$$

and $c = \{c_i\}$ ($i = 1, \dots, 6$) for the three cubic lattices are

$$\begin{aligned} c &= (1.3047, 0.2305, 0.4054, 0.07231, 0.1526, 0.0105) , \\ c &= (0.129, -0.41286, 0.76421, 0.2569, 0.0113, 0.00562) , \\ c &= (0.07529, 0.69657, -0.741, 0.04195, 0.0231, 9.14 \times 10^{-7}) , \end{aligned}$$

for the SC, BCC, and FCC lattices, respectively. Batchelor and O'Brien (1977) considered the problem of calculating the conductivity of a lattice of *touching* spheres with finite conductivity, and derived the following expression for the conductivity of a SC array of touching spheres,

$$g_e \approx \pi \ln \sigma , \quad \sigma \gg 1 . \quad (9.65)$$

The predictions of McPhedran and McKenzie (1978) agree with those of Eq. (9.65) for large values of σ .

McKenzie *et al.* (1978) analyzed the conduction problem in the BCC and FCC lattices. In the case of a BCC lattice, the volume fraction of the spheres is $\phi_2 = 8\pi R^2/3$. The spheres begin to touch at $R = \sqrt{3}/4$. In addition to the exact solution, an equation similar to Eq. (9.58) was derived with $a_1 = 0.057467$, $b_1 = 0.166117$, $b_2 = 1.35858$, $c_1 = 0.000950738$, $c_2 = 0.733934$, $c_3 = 0.134665$, and $c_4 = 0.0465862$. In this case, ϕ_2 must be relatively small in order for the expression to be accurate. The Zuzovsky–Brenner expression, Eq. (9.60), also provides accurate estimates of the effective conductivity with $d_1 = -0.0897$ and $d_2 = 0.03811$.

In a similar manner, an exact expression for the conductivity of a dispersion of equal-size spheres inserted at the nodes of a FCC lattice in a uniform matrix was derived. In this case, the volume fraction of the spheres is $\phi_2 = 16\pi R^3/3$, so that the critical volume fraction at which the spheres begin to touch (i.e., at $R = 1/(2\sqrt{2})$) is $\phi_{\max} = \sqrt{2}\pi/6 \simeq 0.740$. Once again, the exact solution was derived, and an accurate approximant similar to Eq. (9.58) was also derived for low values of the spheres volume fraction ϕ_2 with $a_1 = 0.0047058$, $b_1 = 0.130683$, $b_2 = 1.20500$, $c_1 = 0.00603255$, $c_2 = 5.73021$, $c_3 = 8.20884$, and $c_4 = 0.295595$. Equation (9.60) also provides accurate estimates of the effective conductivity with $d_1 = -0.0685$ and $d_2 = -0.03767$.

Similar to the flow problem, the problem of conduction in 2D systems that consist of regular arrays of infinitely long cylinders with circular cross sections inserted in a uniform matrix has been studied by a large number of groups. As in the 3D case, the conductivities of the cylinders (circles) and the matrix are different, with the radius of the cylinders being R . According to the *International Tables for X-Ray Crystallography*, there are five ways of packing cylinders in regular 2D arrays. Two of such arrays, namely, the square and hexagonal arrays, have been analyzed extensively by many groups (Sahimi, 2003a). We summarize the results for the general problem in which both the cylinders and the matrix have finite conductivities, derived by Perrins *et al.* (1979b) whose method is similar to that of McPhedran and McKenzie (1978) and McKenzie *et al.* (1978).

In the case of a hexagonal array of cylinders of radius R , the volume fraction ϕ_2 of the cylinders is given by $\phi_2 = 2\pi R^2/\sqrt{3}$. They touch when $R = 1/2$ and, therefore, the critical volume fraction is $\phi_{\max} = \pi/(2\sqrt{3})$. Perrins *et al.* (1979b)

derived the exact solution of the problem and, by truncating the exact solution, obtained the following expression for the limit in which the conductivity ratio σ is small, or if the cylinders are not too close to each other:

$$g_e = 1 - 2\phi_2 \left(\mathcal{R} + \phi_2 - \frac{0.075422\mathcal{R}\phi_2^6}{\mathcal{R}^2 - 1.060283\phi_2^{12}} - \frac{0.000076\phi_2^{12}}{\mathcal{R}} \right)^{-1}, \quad (9.66)$$

where $\mathcal{R} = (1 + \sigma)/(1 - \sigma)$. In the case of a square array of cylinders (Perrins *et al.*, 1979a), the critical volume fraction at which the cylinders begin to touch is $\phi_{\max} = \pi/4$. An accurate truncated solution obtained from the exact expression is given by

$$g_e = 1 - 2\phi_2 \left(\mathcal{R} + \phi_2 - \frac{0.305827\mathcal{R}\phi_2^4}{\mathcal{R}^2 - 1.402948\phi_2^8} - \frac{0.013362\phi_2^8}{\mathcal{R}} \right)^{-1}. \quad (9.67)$$

The limit in which the cylinders are in contact is also of interest. Asymptotic analysis of O'Brien (1979) indicates that for a pair of cylinders of conductivity σ that are separated by a small gap h , the current $I(\sigma, h)$ flowing in response to a unit applied field is given by

$$I(\sigma, h) \simeq \left(\frac{2}{\pi} \frac{\ln \sigma}{\sigma} + \frac{1}{\pi} \sqrt{\frac{h}{R}} \right)^{-1}, \quad \sigma \gg 1. \quad (9.68)$$

For a square array of cylinders, I is also the total current flowing in the unit cell if the contribution to g_e from the region around the touching line is much larger than that from other regions. For a hexagonal array of cylinders, I , as given by Eq. (9.68), must be multiplied by $\sqrt{3}$ in order to obtain g_e . In the limit of touching cylinders ($h = 0$), one obtains

$$g_e \simeq \begin{cases} \frac{\pi}{2} \frac{\sigma}{\ln \sigma}, & \text{square array,} \\ \frac{\sqrt{3}\pi}{2} \frac{\sigma}{\ln \sigma}, & \text{hexagonal array,} \end{cases} \quad (9.69)$$

which agree with the calculations of Perrins *et al.* (1979a,b) if $\sigma > 80$. Also, see Milton *et al.* (1981) for arrays of intersecting square prisms and comparison with experimental data. Finally, Harter and Knudby (2004) computed the effective conductivity of periodic arrays of cuboid inclusions located on an octahedral lattice.

9.4

Effective-Medium and Mean-Field Approximations

The exact solutions presented in the last section are elegant, but useful only for certain simplified microstructures. They generally have very little utility for *disordered* porous media. Thus, various approximations have been developed in order to estimate the flow and transport properties of disordered porous media. In this section, one such approximation, the effective-medium approximation (EMA), is

described. It is a popular approximation that provides the correct qualitative trends for the flow and transport properties as functions of the porosity, but is inaccurate under a variety of conditions.

9.4.1

Fluid Flow

An EMA was first developed by Brinkman (1947), although he did not refer to his results as such. Consider a cylindrical column of length L that is packed isotropically with a distribution of identical spheres of radius R . The flow is along the cylinder's axis, say the z direction. A pressure gradient ∇P is applied to the porous medium. We assume for now that the viscosity is unity. We then embed a spherical test particle of radius R in the porous medium and assume that, far from it, the fluid velocity is $\mathbf{V} = V_0 \mathbf{e}_z$, where $\mathbf{e}_z$ is the unit vector. The question that Brinkman asked was, what is the governing equation for the fluid flow? Brinkman suggested that

$$\nabla^2 \mathbf{V} - \frac{1}{K_e} \mathbf{V} = \nabla P, \quad r > R, \quad (9.70)$$

which should be coupled to the continuity equation and no-slip boundary condition at $r = R$. If there is no solid particle, then $K_e \rightarrow \infty$, and one recovers the usual Stokes' equation. On the other hand, in a porous medium in which l is a typical pore length scale, the first term on the left side of Eq. (9.70) is $\mathcal{O}(\tilde{V}/L^2)$, where $\tilde{V}$ and L are the typical macroscopic velocity and length scales. The second term on the left side, on the other hand, is $\mathcal{O}(\tilde{V}/K_e)$ or $\mathcal{O}(\tilde{V}/l^2)$. Because $L \gg l$, the first term can be neglected and one recovers the Darcy's law. Therefore, Brinkman's equation interpolates between the microscopic Stokes' equation and the macroscopic Darcy's law.

The solution of Brinkman's equation is given by

$$\mathbf{V}(\mathbf{r}) = \nabla \left\{ \left[V_0 r + c_1 \frac{(1 + c_2 r) \exp(-c_2 r) - 1}{c_2^2 r^2} - \frac{c_3}{r^2} \right] \cos \theta \right\} + \frac{c_1}{r} \exp(-c_2 r) \mathbf{e}_z, \quad (9.71)$$

$$P = \left(-c_2^2 V_0 r + \frac{c_1}{r^2} + \frac{c_2^2 c_3}{r^2} \right) \cos \theta, \quad (9.72)$$

where θ is the spherical polar coordinate with a polar axis in the positive z direction. The three constants are given by

$$c_1 = -\frac{3}{2} V_0 R \exp(c_2 R), \quad c_2 = K_e^{-\frac{1}{2}},$$

$$c_3 = \frac{1}{2} V_0 R \left[-R^2 + \frac{3}{c_2^2} (e^{c_2 R} - 1 - c_2 R) \right].$$

Note that the exponential term that contains the $1/\sqrt{K_e}$ term indicates that $\sqrt{K_e}$ represents a *screening length*. The drag force F on all the particles is then given by

$$F = 6\pi R V_0 N \left(1 + \frac{R}{\sqrt{K_e}} + \frac{R^2}{3K_e} \right), \quad (9.73)$$

where N is the number of particles. The force F should also be equal to $S\Delta P$, where S is the cross-sectional area of the porous medium perpendicular to the direction of macroscopic flow. Thus, solving for ΔP in terms of the Darcy velocity V_0 and the effective permeability K_e , we obtain an expression for K_e :

$$\frac{K_s}{K_e} = 1 + \frac{R}{\sqrt{K_e}} + \frac{R^2}{3K_e}, \quad (9.74)$$

where $K_s = 2R^2/(9\phi_2)$ is the Stokes' permeability, valid for large porosities (see the next section). Thus, to leading order in ϕ_2 ,

$$\frac{K_s}{K_e} = 1 + \frac{3}{\sqrt{2}}\phi_2^{1/2}. \quad (9.75)$$

Note that according to Eq. (9.75), K_e vanishes at $\phi_2 = 2/3$, that is, when the porosity is $\phi = 1 - \phi_2 = 1/3$.

In his original formulation, Brinkman (1947) assumed that $\mu \neq 1$. Thus,

$$\mu \nabla^2 \mathbf{v} - \frac{\mu}{K_e} \mathbf{v} = \nabla P. \quad (9.76)$$

Koplik *et al.* (1983) suggested, however, that one must use a viscosity μ' in Eq. (9.76) – called the *renormalized viscosity* – which, in principle, is different from μ , the pure-fluid viscosity used in Eq. (9.1). Note that, similar to K_e , the ratio μ'/μ is a property of porous media, and may be related to a phenomenological *slip velocity* parameter that was identified experimentally by Beavers and Joseph (1967) and justified theoretically by Saffman (1971). For an unconsolidated porous medium modeled by a random collection of nonoverlapping spherical particles, Koplik *et al.* (1983) suggested that for small ϕ_2 , one has

$$\frac{\mu'}{\mu} = 1 - \frac{1}{2}\phi_2. \quad (9.77)$$

However, the problem of estimating μ' at high values of ϕ_2 – that is, at low porosities – has not been solved yet. Wilkinson (1985) assumed that $\mu' = \mu$, and derived a nonlinear differential equation for K_e . His equation can be integrated numerically if the porosity and grain size distribution are specified; see also Martys *et al.* (1994).

9.4.2

Transport

Several EMA and mean-field approximations (i.e., approximations that ignore the fluctuations in the quantities) have been derived in the past. They are described in detail by Sahimi (2003a). Therefore, only a summary of them is presented here. Consider a large sphere of radius R_0 that itself is composed of N smaller particles of radius R , conductivity g_1 , and volume fraction, $\phi_2 = N(R/R_0)^d$, all in a matrix of conductivity g_2 , where d is the spatial dimension of the medium. We assume that ϕ_2 is small enough that the interaction between the particles is negligible. Then,

the Maxwell (1873)³⁾ or the Maxwell-Garnett (1904)⁴⁾ approximation is given by

$$\frac{g_e - g_1}{g_e + (d-1)g_1} = \phi_2 \left[\frac{g_2 - g_1}{g_2 + (d-1)g_1} \right]. \quad (9.78)$$

The approximation is useful only when ϕ_2 is small. It can be generalized to the case in which the particles are unidirectionally aligned isotropic ellipsoidal inclusions:

$$\sum_{j=1}^2 \phi_j (g_e - g_i) \cdot \mathbf{R}_{j1} = 0, \quad (9.79)$$

where $\phi_1 = \phi$ is the porosity, $g_i = g_i \mathbf{I}$, and

$$\mathbf{R}_{j1} = \left(\mathbf{I} + \mathbf{A}^* \frac{g_j - g_1}{g_1} \right)^{-1}. \quad (9.80)$$

$\mathbf{A}^*$ is called the *depolarization tensor* that, in the principal axes frame, has diagonal components (or eigenvalues), A_i^* , $i = 1, 2$, and 3 , given by

$$A_i^* = \left(\prod_{i=1}^d \frac{1}{2} a_i \right) \int_0^\infty \frac{dz}{(z + a_i^2) \sqrt{\prod_{i=1}^d (z + a_i^2)}}, \quad (9.81)$$

where a_i is the semi-axis of the ellipsoid along the x_i direction. For d -dimensional spheres, we have $A_i^* = 1/d$.

In the EMA originally derived by Bruggeman (1935) and independently by Landauer (1952), the disordered medium is replaced by an effective homogeneous medium with effective conductivity g_e . A spherical inclusion is carved out of the effective medium and is replaced by a conductivity g , which is distributed according to the statistical distribution of the conductivities in the original disordered medium. The single inclusion produces a perturbation in the potential field throughout the effective medium. g_e is then computed by requiring the average of the perturbations to be zero, with the averaging taken with respect to the statistical distribution of the inclusion's conductivity. Thus, if the original disordered medium consists of n phases with conductivities $g_1, \dots, g_n$ and volume fractions $\phi_1, \dots, \phi_n$, then the EMA predicts that the effective conductivity is the solution of

$$\sum_{i=1}^n \phi_i \frac{g_i - g_e}{g_i + (d-1)g_e} = 0. \quad (9.82)$$

- 3) *James Clerk Maxwell* (1831–1879), a Scottish theoretical physicist and mathematicians, made seminal contributions to electromagnetism. Through Maxwell's equations, he unified electricity, magnetism and light, an achievement referred to as the *second great unification*, after the first one by Isaac Newton.

- 4) *James Clerk Maxwell Garnett* (1880–1958) was a physicist named after J.C. Maxwell because

his father, William Garnett, was Maxwell's co-biographer. He published his theory in 1904, followed by another paper in 1906, but never published a physics paper again. He turned to politics, eventually serving as the Secretary-General of the League of the Nations, the predecessor of the United Nations between World Wars I and II.

The EMA is a popular method for estimating the conductivity. It is reasonably accurate under a variety of conditions, but also not so under other conditions. In fact, Torquato and Hyun (2001) identified single-scale periodic dispersions for which the EMA is *exact* in $d = 2$ for *all* volume fractions of the pore phase. The EMA can also be generalized to the case in which the inclusions are ellipsoidal. In that case,

$$\sum_{j=1}^n \phi_j (\mathbf{g}_j - \mathbf{g}_e) \cdot \mathbf{R}_{je} = 0, \quad (9.83)$$

with

$$\mathbf{R}_{je} = \left[\mathbf{I} + \mathbf{A}^* \cdot \mathbf{g}_e^{-1} \cdot (\mathbf{g}_j - \mathbf{g}_e) \right] = 0. \quad (9.84)$$

Finally, we must also mention the *differential* EMA, also introduced by Bruggeman (1935). In this approximation,

$$\left(\frac{g_2 - g_e}{g_2 - g_1} \right) \left(\frac{g_1}{g_e} \right)^{\frac{1}{d}} = \phi = 1 - \phi_2. \quad (9.85)$$

In this approximation, the pore space remains connected at all the porosities. This particular feature enables one to derive the well-known Archie's law which is used extensively for sedimentary rock. We will come back to the EMA and Archie's law in Chapter 10. See, for example, Schwartz *et al.* (1995) for the application of the differential EMA to evaluating the effective conductivity of mortar, a composite of inert sand grains surrounded by a porous cement paste matrix.

9.5 Cluster Expansion

In the early works on flow and transport in disordered porous media, the *dilute approximations* were made. That is, it was assumed that the porosity is large. In the context of models of porous media in terms of packing of particles, for example, the assumption implies that the volume fraction of the particles is small, so that the interactions between them could be neglected. Then, the approximations were systematically improved by taking into account the interactions between two particles, three particles, and so on. The systematic improvements are called *cluster expansions* since one considers the interactions between the particles in a cluster of size 2, 3, ...

9.5.1 Fluid Flow

First, consider a dilute bed of spherical particles of radius R . Since the spheres do not interact with each other, only the applied pressure ∇P_0 and the drag force $\mathbf{f}$ suffice to compute the effective permeability. However, from the classical solution

for the slow flow of a Newtonian fluid around a single sphere (recall that, in the dilute limit, the interactions between the spheres are neglected), we have $\mathbf{f} = 6\pi\mu R\mathbf{v}$, where $\mathbf{v}$ is the fluid velocity and, $\nabla P_0 = -\rho_p \mathbf{f}$, with ρ_p being the number density of the particles. Thus, using the expression for the pressure gradient in Eq. (9.1), we obtain

$$K_e = \frac{2R^2}{9\phi_2}, \quad (9.86)$$

which is the Stokes' permeability K_s mentioned earlier. If the spheres do not have the same sizes, then Eq. (9.86) is modified to

$$K_e = \frac{2\langle R^3 \rangle}{9\langle R \rangle \phi_2}. \quad (9.87)$$

Equation (9.86) has been generalized to the case in which one has a dilute bed of spheroids (Torquato, 2002). The effective permeability is given by

$$K_e = \frac{2a^2}{9\phi_2} \begin{bmatrix} f\left(\frac{b}{a}\right) & 0 & 0 \\ 0 & f\left(\frac{b}{a}\right) & 0 \\ 0 & 0 & g\left(\frac{b}{a}\right) \end{bmatrix}, \quad (9.88)$$

where the spheroid's semi-axes are $a_1 = a_2 = a$ and $a_3 = b$, and $\phi_2 = (4/3)\pi\rho_p a^2 b$, with ρ_p being the number density of the spheroids. The function $f(b/a)$ is given by

$$f\left(\frac{b}{a}\right) = \begin{cases} \frac{3}{16\chi_b^3} \left[(3\chi_b^2 - 1) \ln\left(\frac{1+\chi_b}{1-\chi_b}\right) + 2\chi_b \right], & \frac{b}{a} \geq 1, \\ \frac{3}{8\chi_a^3} \left[(3\chi_a^2 + 1) \tan^{-1} \chi_a - \chi_a \right], & \frac{b}{a} \leq 1, \end{cases} \quad (9.89)$$

while

$$g\left(\frac{b}{a}\right) = \begin{cases} \frac{3}{8\chi_b^3} \left[(\chi_b^2 + 1) \ln\left(\frac{1+\chi_b}{1-\chi_b}\right) - 2\chi_b \right], & \frac{b}{a} \geq 1, \\ \frac{3}{4\chi_a^3} \left[(\chi_a^2 - 1) \tan^{-1} \chi_a + \chi_a \right], & \frac{b}{a} \leq 1. \end{cases} \quad (9.90)$$

Here, $\chi_a^2 = -\chi_b^2 = a^2/b^2 - 1$.

Childress (1972), Howells (1974), and Hinch (1977) studied flow through a *nondilute* and *random* array of spheres, and obtained an asymptotic expression for the permeability of the porous medium for low values of ϕ_2 (high porosities). Hinch (1977)'s result is given by

$$\frac{K_s}{K_e} = 1 + \frac{3}{\sqrt{2}} \phi_2^{\frac{1}{2}} + \frac{135}{64} \phi_2 \ln \phi_2 + 16.456 \phi_2 + o(\phi_2). \quad (9.91)$$

Actually, his result contained a few numerical errors that were corrected by Kim and Russel (1985), who also derived a few higher order terms of expansion (9.91).

9.5.2

Transport

We only consider two limiting cases in the nondilute case, and provide the expressions for the effective conductivity. If the spheres are not penetrable (see Chapter 6), then Jeffrey (1973) showed that

$$\frac{g_e}{g_1} = 1 - \frac{3}{2}\phi_2 + 0.588\phi_2^2 + \mathcal{O}(\phi_2^3) , \quad (9.92)$$

whereas if the spheres are fully penetrable (the Swiss-cheese model; see Chapter 3), then (Torquato, 2002; Sahimi, 2003a)

$$\frac{g_e}{g_1} \simeq 1 + 3\phi_2 + 7.556\phi_2^2 + \mathcal{O}(\phi_2^3) . \quad (9.93)$$

In both cases, g_1 represents the conductivity of the “matrix”, representing that of brine in the case of porous media.

9.6

Rigorous Bounds

As the discussions so far make it abundantly clear, it is, in general, impossible to exactly determine the effective flow and transport properties of *disordered* porous media. Thus, the most rigorous statements and results for such properties are in terms of upper and lower bounds. Considerable progress has been made over the past three decades in deriving such bounds for the effective permeability, conductivity, and other properties of the continuum models. As Torquato (2002) pointed out, the bounds are useful because

1. They incorporate rigorously nontrivial information about the microstructure of the system;
2. as more microstructural information is included, the gap between the upper and lower bounds becomes narrower;
3. one of the bounds (either the lower or upper bound) usually provides an accurate estimate of the property of interest for a range of conditions;
4. the bounds become *exact* for certain microstructures, and
5. they can be used to test the merit of an approximation or theory.

9.6.1

Fluid Flow

Prager (1961, 1963) and Weissberg and Prager (1962, 1970) pioneered derivation of the upper and lower bounds to K_e . (Note that Berryman and Milton (1985) corrected an error in the original results of Weissberg and Prager.) They proposed that certain

bounds on K_e , which depend upon various distribution functions that statistically characterize the porous medium, may be used for estimating K_e for a wide range of sphere volume fraction ϕ_2 , or the porosity $\phi = 1 - \phi_2$. Weissberg and Prager (1962, 1970) evaluated the bounds for a model in which the centers of the spheres are distributed at random, the so-called *fully-penetrable sphere model*, or the Swiss-cheese model described in Chapter 6.

Torquato and Beasley (1987) considered the general case in which the spheres have an arbitrary degree of impenetrability characterized by a parameter λ , where $\lambda = 0$ corresponds to the fully-penetrable model, whereas $\lambda = 1$ represents the case of completely-impenetrable spheres (see Chapter 6). They derived useful upper and lower bounds for the permeability of the porous medium as a function of λ . We refer the reader to Torquato (2002) for more details and bounds, and summarize the sharpest bounds. For the fully-penetrable sphere model (the Swiss-cheese model) with identical spheres, the lower bound of Weissberg and Prager is given by

$$\frac{K_s}{K_e} \geq -\frac{\ln \phi}{\phi \phi_2} . \quad (9.94)$$

For *dilute* random arrays of identical spheres in the penetrable-concentric-shell model (the cherry-pit model described in Chapter 6), Torquato and Beasley (1987) derived the following lower bound

$$\frac{K_s}{K_e} \geq 1 + \left[\frac{3}{2} - \lambda^6 + \frac{11}{4} \lambda^4 + \frac{5}{6} \lambda^2 - \frac{9}{8} \lambda + \frac{3}{4} (1 + 3\lambda^2) \ln(1 + 2\lambda) - \frac{\lambda(1 + 7\lambda)}{16(1 + 2\lambda)^2} \right] \phi_2 , \quad (9.95)$$

which is a relatively accurate bound for $0.52 \leq \lambda \leq 1$. However, for $\lambda \leq 0.52$, they derived the following lower bound

$$\frac{K_s}{K_e} \geq 1 + \frac{5}{8}(3 + 5\lambda) + \mathcal{O}(\phi_2^2) , \quad (9.96)$$

which is a more accurate bound than (9.94). Note that bounds (9.95) and (9.96) can be written in terms of ϕ , the porosity of the system, using the relations between ϕ and ϕ_2 described in Chapter 6. For a model of polydispersed and overlapping spheres with radii that follow the Schulz distribution

$$f(R) = \frac{1}{\Gamma(\alpha + 1)} \left(\frac{\alpha + 1}{\langle R \rangle} \right) R^2 \exp \left[-\frac{(\alpha + 1)R}{\langle R \rangle} \right] , \quad \alpha > -1 , \quad (9.97)$$

where $\Gamma(x)$ is the gamma function, and α is a measure of polydispersivity ($\alpha = \infty$ corresponds to monodispersed spheres), Torquato and Lu (1990) calculated a lower bound to K_e . Their results are shown in Figure 9.2. An interesting result of Torquato and Beasley (1987) was that the bounds that incorporate a certain level of statistical information on the medium are *not* necessarily more accurate than those that incorporate *less* information. It should be mentioned that the Kozeny–Carman empirical formula

$$\frac{K_s}{K_e} = \frac{10\phi_2}{(1 - \phi_2)^3} , \quad (9.98)$$

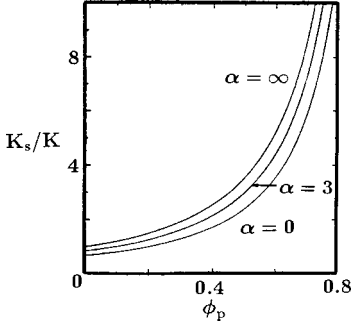


Figure 9.2 Lower bounds on the permeability K_e of a model of polydispersed and overlapping spheres, with radii that follow the Schulz distribution versus the spheres' volume fraction ϕ_2 . $\alpha = \infty$ corresponds to a monodispersed system (after Torquato and Lu, 1990).

is relatively close to the lower bound for the permeability of the random sphere model. For a comprehensive discussion of the origin and merits of Eq. (9.98), see Scheidegger (1974).

9.6.2

Transport

Rigorous and relatively accurate bounds have been derived for a variety of continuum models. The reader should consult Torquato (2002) and Sahimi (2003a) for detailed derivation, discussions, and merit of such bounds.

9.7

Empirical Correlations

There are several empirical or semi-empirical correlations for representing the relation between the pressure drop and the fluid velocity in packings of particles. One of the best-known of such correlations is the *Blake–Kozeny* equation

$$V_s = \frac{1}{c_k} \frac{\Delta P}{L} \frac{D_p^2}{36\mu} \frac{\phi^3}{(1-\phi)^2}, \quad (9.99)$$

where V_s is the superficial velocity of the fluid, which is related to the mean flow velocity V_m by $V_s = V_m\phi$. Here, c_k is called the Kozeny constant with a numerical value of about five, and D_p is an equivalent particle diameter defined by

$$D_p = \frac{6}{\Xi}, \quad (9.100)$$

where Ξ is the specific surface area of the particles. Equation (9.99) is used at low Reynolds numbers, where the inertial effects are not important. If such effects are important, then the *Blake–Plummer* equation is combined with the Blake–Kozeny

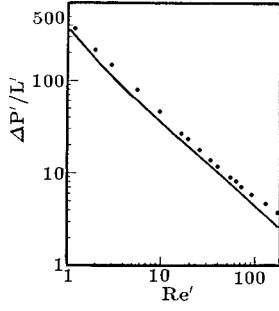


Figure 9.3 Comparison of the experimental (circles) and calculated (curve) pressure drops for flow through an in-line bundle of tubes (after Eidsath *et al.*, 1983).

equation, with the former being given by

$$V_s^2 = \frac{4}{7} \frac{\Delta P}{L} \frac{D_p}{\rho} \frac{\phi^3}{1-\phi}, \quad (9.101)$$

with ρ being the density of the fluid. If Eqs. (9.99) and (9.101) are simply added together, one obtains the *Ergun* equation,

$$\frac{\Delta P}{L} = \frac{36c_k\mu}{D_p^2} \frac{(1-\phi)^2}{\phi^3} V_s + \frac{7\rho}{4D_p^2} \frac{1-\phi}{\phi^3} V_s^2, \quad (9.102)$$

which interpolates between the two equations.

MacDonald *et al.* (1979) reviewed the pressure drop-flow rate data for a large number of unconsolidated porous media and suggested that if the particles are smooth, the data can be represented by the following equation

$$\frac{\Delta P'}{L'} = \frac{180}{Re'} + 1.8, \quad (9.103)$$

with

$$\Delta P' = \frac{\Delta P}{\rho V_s^2}, \quad L' = \frac{L}{D_p} \frac{1-\phi}{\phi}, \quad Re' = \frac{\rho V_s D_p}{\mu} \frac{\phi}{1-\phi}.$$

Typical experimental data that follow Eq. (9.103) are shown in Figure 9.3. Another popular empirical correlation is the *Kozeny–Carman* equation, Eq. (9.98). Its predictions fall within 15% of the results for at least one of the three types of periodic packings described earlier if $\phi_2 > 0.5$ and, in the case of a random packing of spheres, they are relatively close to the lower bound given above. Many other empirical and semi-empirical correlations have been suggested in the past, a list of which would be too long to be given here. Also, see Martins *et al.* (2007), who provide extensive references to older works on flow through packed beds.

9.8

Packings of Nonspherical Particles

Since many natural porous media, for example, sandstones, are not composed of spherical particles, it is important to study the effect of particle shape on the

permeability and transport properties. The problem has been studied extensively. Schwartz and Banavar (1989) developed a model in which one first constructs a packing of spherical particles. The centers of the spherical particles were then fixed, while their shapes were deformed into ellipses. The effective permeability was then estimated by an EMA (see also Chapter 10), and was shown to be strongly influenced by the particles' shape. In an extension of the work, Pilotti (1998) deformed the surface of the spheres in a packing in order to model real aspherical grains in a sandstone.

Coelho *et al.* (1997) carried out computer simulation of settling of monosized aspherical particles, such as, ellipsoids and parallelepipeds. It was shown that in the porosity range $0.4 < \phi < 0.8$, the packings of different particles, but with the same porosity, had similar hydraulic properties, regardless of the particles' shape. Similar conclusions were reached by Lehmann *et al.* (2008), who studied packings of overlapping ellipsoids.

Koponen *et al.* (1997) constructed packings of square-shape particles and estimated their permeability by computer simulations. They also presented a modification of the Kozeny's equation, and proposed an analytical expression for the specific surface area of the packings.

Garcia *et al.* (2009a) used numerical simulations to study the dependence of the permeability of packings of particles on the particle shape and size distribution. Their results indicated that the details of particle shape and size distribution have only a small effect on the permeability of the packings. Garcia *et al.* (2009b) suggested a method for constructing irregular particles based on images of the real particles obtained by, for example, scanning. The particle's shape is defined by a surface mesh. Then, spheres are added sequentially to the volume enclosed by the mesh. Adding more spheres generates a cluster that increasingly resembles the surface of the real particle. There is also a library of shapes of real particles that were obtained by scanning (Latham *et al.*, 2008).

9.9

Numerical Simulation

Numerical simulation of flow and transport through packed beds can be carried by several methods. The main problem in such simulations is the curved boundaries at the fluid-solid interface. One of the most heavily-used methods is the Galerkin finite-element method as it provides a flexible and accurate technique for handling such complex boundaries. Over the years, various versions of the method have been used for simulating flow through packing of particles; see, for example, Sørensen and Stewart (1974), Zick and Homsy (1982), and Eidsath *et al.* (1983) all the way to Garcia *et al.* (2009a), who provide more references.

Another method is to map the packing onto an equivalent network of bonds and sites, with appropriate assignments of the volumes of the sites and the bonds. If this mapping can be done accurately, then network simulation techniques that are described in Chapter 10 can be used for calculating the effective permeability and

other properties of such model porous media. Such a network model was used by Bryant *et al.* (1993a,b), Thompson and Fogler (1997), Øren and Bakke (2002), Dadvar and Sahimi (2003), and Martins *et al.* (2007) in their simulations of flow and transport through packed beds of particles.

One may take advantage of the approximate (but accurate) relation between the permeability and conductivity of a porous medium proposed by Johnson *et al.* (1986), which is described below. Since the relation between the two has been shown to be accurate for packings of particles, one can first calculate the electrical conductivity of the system by a random walk method (see below) which can be done efficiently and without any need for solving any equation explicitly, and then use the relation between the permeability and electrical conductivity to estimate K_e . Other more recent methods for numerical simulation of flow and transport through packings of particles have been described in considerable detail by Torquato (2002) and Sahimi (2003a).

9.9.1

Random Walk Methods

It has been known for decades (see, for example, Chandrasekhar, 1943⁵; Hughes, 1995) that from the time-dependence of the mean-square displacement $\langle R^2(t) \rangle$ of a random walker that diffuses in a heterogeneous medium, one can compute the effective diffusivity D_e and conductivity g_e . In particular, the effective conductivity of a d -dimensional medium is given by

$$g_e = \lim_{t \rightarrow \infty} \frac{\langle R^2(t) \rangle}{2dt}, \quad (9.104)$$

where t is the time and $\langle \cdot \rangle$ denotes an ensemble average. A similar equation also holds for the effective diffusivity D_e . The first application of this idea to determining the effective conductivity of composite materials appeared in a paper by Haji-Sheikh and Sparrow (1966), who studied heat conduction in a composite solid. Over the past four decades, random walk methods have been used extensively for studying transport in disordered media. Hughes (1995) provides an extensive review of fundamental properties of random walks. In this section, we describe the application of this technique for determining the effective conductivity (and, hence, diffusivity) of porous and composite media that are represented by the continuum models, while in 10, the implementation of the method in the pore network models of porous media will be described.

The essential idea is simple: One computes the mean-square displacement of some random walkers in the system and utilizes Eq. (9.104) to estimate g_e . The simulations are sped up by using the concept of *first-passage time* (FPT), which is

- 5) Subrahmayan Chandrasekhar (1910–1995) was an Indian-American astrophysicist who, along with William Alfred Fowler (1911–1995), was awarded the Nobel Prize for Physics in 1983 for his seminal work on the evolution of the structure of stars. He was a nephew of Chandrasekhara Venkata Raman (1888–1970), who was awarded the same Prize in 1930 for his discovery of Raman scattering.

the statement of the fact that if a random walker moves in a homogeneous region of a material (for example, within one phase of a two-phase material where the local conductivity is essentially the same everywhere within that phase), there is no need to spend unnecessary time to simulate its motion in detail. Instead, the walker should be allowed to take long steps to quickly pass through the homogeneous regions and arrive at the interface between the two phases. The necessary time for taking the long steps can often be calculated *analytically*, hence resulting in further improvement in the efficiency of the method. The only requirement for using the FPT method is that the walker should not take such long steps that would take it outside of a homogeneous region. After taking each of such long steps, the time is increased by an amount appropriate to that step.

To our knowledge, the FPT technique was first used by Sahimi *et al.* (1982) for simulating hydrodynamic dispersion (to be studied in Chapters 11 and 12) in flow through a porous medium. In the context of calculating the effective conductivity and diffusivity of a disordered material, Siegel and Langer (1986) and Kim and Torquato (1990) appear to be the first to have used the method. Let us now describe the details of the FPT method for calculating the effective conductivity (and hence diffusivity) of disordered materials.

We first consider a homogeneous medium and follow Torquato *et al.* (1999) to describe the essentials of the FPT method. Suppose that a random walker is diffusing in a d -dimensional homogeneous material of conductivity g . We surround the particle with a *first-passage region* Ω that has a bounding surface $\partial\Omega$, and denote by $\mathbf{r}$ and $\mathbf{r}_B$ the position of the walker inside Ω and on its boundary, respectively. Let $P(\mathbf{r}, \mathbf{r}_B, t)$ be the probability associated with the walker hitting $\partial\Omega$ in the vicinity of $\mathbf{r}_B$ for the *first time* at time t if it starts its motion at $\mathbf{r}$. $P(\mathbf{r}, \mathbf{r}_B, t)$ is a probability density function in the variable $\mathbf{r}_B$ and a cumulative probability distribution function in the time variable t that satisfies the diffusion equation:

$$g\nabla^2 P(\mathbf{r}, \mathbf{r}_B, t) = \frac{\partial P(\mathbf{r}, \mathbf{r}_B, t)}{\partial t}, \quad (9.105)$$

subject to the boundary and initial conditions that

$$P(\mathbf{r}, \mathbf{r}_B, t = 0) = 0, \quad \mathbf{r} \in \Omega \quad (9.106)$$

$$P(\mathbf{r}, \mathbf{r}_B, t) = \delta(\mathbf{r} - \mathbf{r}_B), \quad \mathbf{r} \text{ on } \partial\Omega, \quad t > 0. \quad (9.107)$$

Now, suppose that $C(\mathbf{r}, t)$ is the cumulative probability distribution function associated with the walker starting at $\mathbf{r}$ to first hit any point on the surface $\partial\Omega$ at time t . Clearly, $C(\mathbf{r}, t)$ is the integral of $P(\mathbf{r}, \mathbf{r}_B, t)d\mathbf{r}_B$, integrated over $\partial\Omega$. Hence, it is not difficult to show that $C(\mathbf{r}, t)$ also satisfies the same unsteady-state diffusion equation as $P(\mathbf{r}, \mathbf{r}_B, t)$ with the same initial condition, but with the boundary condition that $C(\mathbf{r}, t) = 1$ for $\mathbf{r}$ in $\partial\Omega$.

An important physical quantity is the *average hitting time* of the average FPT, $t_h(\mathbf{r})$, the average time that a random walker takes, starting from $\mathbf{r}$, to hit the surface

$\partial\Omega$ for the *first time*. It is not difficult to see that

$$t_h(\mathbf{r}) = \int_0^\infty \frac{\partial C}{\partial t} dt . \quad (9.108)$$

In particular, we are interested in the average hitting time t_0 when the walker begins its walk from the origin, $t_0 = t_h(\mathbf{0})$. For example, for a d -dimensional sphere of radius R centered at the origin, one has $t_0 = R^2/(2dg)$, where g is the sphere's conductivity. In principle, one can solve the unsteady-state diffusion equation for $C(\mathbf{r}, t)$ from the solution of which the average hitting time can be computed. However, Torquato *et al.* (1999) showed that t_h satisfies the following *steady-state* diffusion equation

$$g\nabla^2 t_h(\mathbf{r}) = -1 , \quad \mathbf{r} \in \Omega , \quad (9.109)$$

$$t_h(\mathbf{r}) = 0 , \quad \mathbf{r} \text{ on } \partial\Omega . \quad (9.110)$$

Another important property is the probability density $w(\mathbf{r}, \mathbf{r}_B)$ associated with hitting the vicinity of a particular position $\mathbf{r}_B$ on the surface $\partial\Omega$ for the first time if the walker starts at $\mathbf{r}$. It is not difficult to see that $w(\mathbf{r}, \mathbf{r}_B) = \int_0^\infty \partial P/\partial t dt = P(\mathbf{r}, \mathbf{r}_B, t = \infty)$, hence implying that $w(\mathbf{r}, \mathbf{r}_B)$ satisfies the following boundary-value problem,

$$\nabla^2 w(\mathbf{r}, \mathbf{r}_B) = 0 , \quad \mathbf{r} \in \Omega , \quad (9.111)$$

$$w(\mathbf{r}, \mathbf{r}_B) = \delta(\mathbf{r} - \mathbf{r}_B) . \quad (9.112)$$

Of particular interest is the probability density $w(\mathbf{r}, \mathbf{r}_B)$ when the walker begins its walk at the origin, $w(\mathbf{r}_B) = w(\mathbf{r} = \mathbf{0}, \mathbf{r}_B)$. A final quantity of interest is the jumping probability $p(\mathbf{r})$, the probability that the random walker, starting at $\mathbf{r}$, arrives on a certain portion of the first-passage surface $\partial\Omega_0$ for the first time. Clearly,

$$p(\mathbf{r}) = \int_{\partial\Omega_0} w(\mathbf{r}, \mathbf{r}_B) d\mathbf{r}_B , \quad (9.113)$$

which implies that $p(\mathbf{r})$ satisfies a steady-state diffusion equation with the boundary condition that

$$p(\mathbf{r}) = \begin{cases} 1 , & \mathbf{r} \text{ on } \partial\Omega_0 , \\ 0 , & \text{otherwise.} \end{cases} \quad (9.114)$$

We now consider a two-phase, d -dimensional heterogeneous material of conductivities g_1 and g_2 . Suppose that the random walker is in the vicinity of the interface between the two phases. We surround the walker with a first-passage region Ω having a bounding surface $\partial\Omega$ that encompasses both phases. Let Ω_i be that portion of Ω that contains phase i (with $i = 1, 2$) with a corresponding surface $\partial\Omega_i$, and Γ be the interface surface. Equation (9.109) is modified to

$$g_i \nabla^2 t_h(\mathbf{r}) = -1 , \quad \mathbf{r} \in \Omega_i , \quad (9.115)$$

subject to the boundary condition (9.110) and the interface conditions

$$t_h|_1 = t_h|_2, \quad \mathbf{r} \text{ on } \Gamma, \quad (9.116)$$

$$\left. \frac{\partial t_h}{\partial n_1} \right|_1 = \frac{g_2}{g_1} \left. \frac{\partial t_h}{\partial n_1} \right|_2, \quad \mathbf{r} \text{ on } \Gamma, \quad (9.117)$$

where n_1 is the unit outward normal from Ω_i . The jump probability $w(\mathbf{r}, \mathbf{r}_B)$ satisfies the same equation and boundary condition as before, but with the additional interface conditions similar to Eqs. (9.116) and (9.117). The probability $p_i(\mathbf{r})$ that the random walker, initially at $\mathbf{r}$, hits the first-passage surface $\partial\Omega_i$ for the first time is governed by an equation similar to Eq. (9.113), but integrated over $\partial\Omega_i$. The boundary conditions for, for example, $p_1(\mathbf{r})$, are given by

$$p_1(\mathbf{r}) = \begin{cases} 1, & \mathbf{r} \text{ on } \partial\Omega_1, \\ 0, & \mathbf{r} \text{ on } \partial\Omega_2. \end{cases} \quad (9.118)$$

Moreover, the interface conditions,

$$p|_1 = p|_2, \quad \mathbf{r} \text{ on } \Gamma, \quad (9.119)$$

$$\left. \frac{\partial p}{\partial n_1} \right|_1 = \frac{g_2}{g_1} \left. \frac{\partial p}{\partial n_1} \right|_2, \quad \mathbf{r} \text{ on } \Gamma, \quad (9.120)$$

must be satisfied. From the obvious relation, $\sum_i p_i(\mathbf{r}) = 1$, one can determine $p_2(\mathbf{r})$ given $p_1(\mathbf{r})$.

Let us demonstrate the FPT method with two examples. Consider a first-passage square with a side length of $2L$ in a homogeneous medium of conductivity g , with the origin of the coordinates being at the square's center. The average hitting time, the solution of Eqs. (9.109) and (9.110), can be easily derived by the method of separation of variables (see, for example, Carslaw and Jaeger, 1959). Then, the average hitting time for a walk starting at the origin is found by setting $\mathbf{r} = \mathbf{0}$ in the solution, yielding

$$t_0 = \frac{L^2}{2g} - \frac{16L^2}{g\pi^3} \sum_{n=0}^{\infty} \frac{(-1)^n}{(2n+1)^3 \cosh\left[(2n+1)\frac{\pi}{2}\right]}. \quad (9.121)$$

Moreover, the series in Eq. (9.121) can be summed numerically, yielding $t_0 \simeq (0.295L^2)/g$. The probability density $w(\mathbf{r}, \mathbf{r}_B)$ is obtained by solving Eq. (9.111) by the method of separation of variables, subject to the boundary conditions that $w(x, -L) = w(x, L) = w(-L, y) = 0$, and $w(L, y) = \delta(y - y_B)$. The solution is given by

$$w(x, y, y_B) = \frac{1}{L} \sum_{n=1}^{\infty} \frac{\sinh\left[\frac{n\pi(x+L)}{2L}\right] \sin\left[\frac{n\pi(y+L)}{2L}\right] \sin\left[\frac{n\pi(y_B+L)}{2L}\right]}{\sinh(n\pi)}, \quad (9.122)$$

from which the probability p that the random walker lands for the first time at any point along the side $x = L$ is determined, using Eq. (9.113), to be $p = 1/4$.

If the first-passage square contains two phases, then depending on how the two phases are distributed in the square, Eqs. (9.121) and (9.122) must be somewhat modified. For example, suppose that the upper and lower halves of the square are occupied by phases 1 and 2, respectively. Then, Eqs. (9.115)–(9.117) must be solved by the method of separation of variables, which is straightforward. Then, setting $r = 0$ yields the following exact result

$$t_0 = \frac{2}{g_1 + g_2} t_{h0} , \quad (9.123)$$

where t_{h0} is the solution for the homogeneous square given by Eq. (9.121) for $g = 1$. It is not difficult to show that Eq. (9.123) is also applicable to the case in which each phase occupies two diagonal quadrants of the square. The jumping probability density function $w(x, y)$ is obtained by solving Eq. (9.111) with the method of separation of variables, subject to the interface and boundary conditions (9.112) and the analogues of Eqs. (9.116) and (9.117) for $w(x, y)$. If we set $x = y = 0$ in the resulting solution and consider the boundary point y_B to be on the $x = L$ side, we obtain

$$w(y_B) = \begin{cases} (2g_2)(g_1 + g_2)^{-1} w_h(y_B) , & -L \leq y_B < 0 , \\ (2g_1)(g_1 + g_2)^{-1} w_h(y_B) , & 0 < y \leq L , \end{cases} \quad (9.124)$$

where $w_h(y_B)$ is the solution for the case of a homogeneous square, Eq. (9.122). Observe that in this case, $w(y_B)$ is discontinuous at $y_B = 0$. The same solution, Eq. (9.124), is obtained if we consider the boundary points y_B to be on the side $x = -L$. The probability p that the random walker lands for the first time along the side $x = L$ is $p = 1/4$. If we consider the boundary point x_B on the $y = L$ side, we obtain

$$w(x_B) = \frac{2g_1}{g_1 + g_2} w_h(x_B) , \quad -L \leq y \leq L . \quad (9.125)$$

The probability p that the random walker lands for the first time at any point along the side $y = L$ is given by

$$p = \int_{-L}^L w(x_B) dx_B = \frac{g_1}{2(g_1 + g_2)} . \quad (9.126)$$

A similar approach is used if the two phases are distributed differently than what we just considered. If, for example, phase 1 occupies three quadrants of the square and phase 2 occupies one quadrant, we obtain

$$t_0 = \frac{4}{3g_1 + g_2} t_{h0} , \quad (9.127)$$

and the corresponding expressions for $w(x_B)$, $w(y_B)$, and p can also be derived.

More generally, suppose that $\Omega = \Omega_1 \cup \Omega_2$ is a small spherical first-passage region of radius R centered at the interface at position $\mathbf{x}_0$, and $\partial\Omega_i$ is the surface of

Ω_i excluding the two-phase interface. One can show (Kim and Torquato, 1992) that for a curved but smooth interface, the probability p_1 (p_2) that the random walker, initially at $\mathbf{x}$ near $\mathbf{x}_0$, eventually arrives on the surface $\partial\Omega_1$ ($\partial\Omega_2$) is given by

$$p_1 = \begin{cases} S_1 (S_1 + \sigma S_2)^{-1} \left(1 + \sigma \sum_{n=0}^{\infty} B_{2n+1} r^{2n+1} \right), & \mathbf{x} \in \Omega_1 \\ S_1 (S_1 + \sigma S_2)^{-1} \left(1 - \sum_{n=0}^{\infty} B_{2n+1} r^{2n+1} \right), & \mathbf{x} \in \Omega_2, \end{cases} \quad (9.128)$$

where

$$B_{2n+1} = \frac{(-1)^n (2n)!}{2^{2n+1} (n!)^2} \frac{4n+3}{n+1}. \quad (9.129)$$

Here, S_i is the area of the surface $\partial\Omega_i$ in phase i , $r = |\mathbf{x} - \mathbf{x}_0|/R$, and $\sigma = g_2/g_1$. The average hitting time t_h for the random walker, initially at $\mathbf{x}$, to hit $\partial\Omega = \partial\Omega_1 \cup \partial\Omega_2$ for the first time is given by

$$t_h = \begin{cases} t_{01} (V_1 + V_2) (V_1 + \sigma V_2)^{-1} \\ \quad \times \left[1 - \frac{r^2(3\sigma-1)}{2} + (\sigma-1) \sum_{n=0}^{\infty} C_{2n+1} r^{2n+1} \right], & \mathbf{x} \in \Omega_1, \\ t_{01} (V_1 + V_2) (V_1 + \sigma V_2)^{-1} \\ \quad \times \left[1 + \frac{r^2(1-3\sigma^{-1})}{2} - (1-\sigma^{-1}) \sum_{n=0}^{\infty} C_{2n+1} r^{2n+1} \right], & \mathbf{x} \in \Omega_2, \end{cases} \quad (9.130)$$

where V_i is the volume of region Ω_i , $t_{01} = R^2/(6g_1)$, and

$$C_{2n+1} = \frac{(-1)^{n+1} (2n)!}{2^{2n+2} (n!)^2} \frac{3(4n+3)}{(n+1)(n+2)(2n-1)}. \quad (9.131)$$

Even if the interface is not smooth, Eqs. (9.128) and (9.130) can still be used by making the sphere radius R smaller.

In practice, in a FPT simulation, one constructs the largest (imaginary) concentric sphere of radius R around a randomly chosen point in phase i , which just touches the multiphase interface. Suppose that the random walker is initially at the center of the imaginary sphere. The average time $t_{0i} = R^2/(2dg_i)$ for the particle to reach a randomly-selected point on the surface of the sphere is recorded, and the process of constructing the sphere and calculating the time a point on its surface is reached is repeated, until the random walker comes within a very small distance of the interface between the two phases. One then computes the average hitting time t_h associated with an imaginary concentric sphere of radius R in the small neighborhood of the interface, and the probability of crossing the interface, both of which were given above. If the random walker crosses the interface and enters a new phase, it finds itself in a new homogeneous phase and, therefore, the

process of constructing imaginary spheres is repeated. The effective conductivity of the heterogeneous material, in the limit $t \rightarrow \infty$, is then given by

$$\frac{g_e}{g_1} = \frac{\left\langle \sum_i t_{01}(R_i) + \sum_j t_{01}(R_j) + \sum_k t_{01}(R_k) \right\rangle}{\left\langle \sum_i t_{01}(R_i) + \sum_j \frac{t_{01}(R_j)}{\sigma} + \sum_k t_h(R_k) \right\rangle}, \quad (9.132)$$

where the summations over i and j are for the random walker's paths in phase 1 and 2, respectively, the summation over k is for the paths that cross the interface boundary, and $\langle \cdot \rangle$ denotes an average over all the realizations of the disordered medium.

The same principles can be used for estimating the effective conductivity tensor g_e of an anisotropic material. In this case, the generalization of Eq. (9.104) for the ij component of the effective conductivity tensor in the limit of long times (or, equivalently, large random walker displacements) is given by

$$(g_e)_{ij} = \frac{x_i x_j}{2t}, \quad (9.133)$$

where x_i is the displacement of the random walker in the i th direction, and t is the *total* time that the random walker spends in order to make a total mean-square displacement $\langle R^2(t) \rangle = \sum_i x_i^2$. The FPT technique can, of course, be used for anisotropic materials as well, and the analogue of Eq. (9.132) for $(g_e)_{ij}$ is given by

$$(g_e)_{ij} = \frac{\langle x_i x_j \rangle}{2 \left\langle \sum_i t_{01}(R_i) + \sum_j t_{01}(R_j) + \sum_k t_h(R_k) \right\rangle}, \quad (9.134)$$

where the notations are exactly the same as in Eq. (9.132).

A random walk method is particularly useful for estimating the electrical conductivity of composite materials made of an insulating matrix and a sample-spanning conducting phase. A well-known example of such systems is a porous medium made of an insulating matrix and a pore space saturated with a conducting fluid. A traditional method, such as the Galerkin finite-element technique, is notoriously time consuming for such materials as a highly resolved finite-element mesh with roughly about one billion nodes would be required to accurately solve the Laplace equation, a prospect which is currently impractical. For this reason alone, a random walk method is the preferred technique for estimating diffusivity and conductivity of heterogeneous materials. Evans *et al.* (1980), Abbasi *et al.* (1983), Nakano and Evans (1983), Akanni *et al.* (1987), and Tomadakis and Sotirchos (1991) used the random walk method to study both ordinary and Knudsen diffusion in a variety of porous media made of random distributions of penetrating or non-penetrating spheres (disks in 2D). Likewise, Smith and Huizenga (1984) used the method to investigate Knudsen diffusion in a random assemblage of spheres.

Schwartz and Banavar (1989) used random walk simulations to calculate the electrical conductivity of the grain-consolidation model of Roberts and Schwartz (1985), described in Chapter 7, with multisize particles. The results, shown in Figure 9.4 in terms of the formation factor $F = g_t/g_e$, are in excellent agreement with the experimental data of Guyon *et al.* (1987) for sintered binary composites, and with other experimental data for similar systems (Oger *et al.*, 1986).

In practice, the grain particles are not usually spherical. However, in a random walk simulation, the particles may have any shape. Schwartz and Banavar (1989) also calculated the electrical conductivities of a porous medium that was originally a packing of spherical rubber grains, but was exposed to a uniaxial pressure that was applied in order to deform the particles. To model the system, they constructed an unconsolidated sphere pack with a given particle distribution, and then compressed the system in one direction by a given α_c . This resulted in a system of spheroidal grains that were then allowed to grow along the three axes. Thus, a sphere of radius R at (x_0, y_0, z_0) was replaced by the spheroid $\alpha_c^{2/3}(x - x_0)^2 + \alpha_c^{2/3}(y - y_0)^2 + \alpha_c^{-4/3}(z - z_0)^2 = R^2$. Note that the resulting system is no longer isotropic and, thus, one must calculate a conductivity tensor. The simulation results of Schwartz and Banavar (1989) were in good agreement with the experimental data of McLachlan *et al.* (1987).

In order to speed up random walk simulations in porous media that have *large-scale* inhomogeneities, such as, layering, Schwartz and Banavar (1989) introduced a weak *bias* in their simulations. The bias causes the random walker to sample the pore space more efficiently because in the direction of the bias, the traveled distance is proportional to N_s , rather than $N_s^{1/2}$, where N_s is the number of the steps. Although the idea of biased diffusion for estimating conductivity of inhomogeneous media is not new, it has been used in the context of random walk simulations only recently.

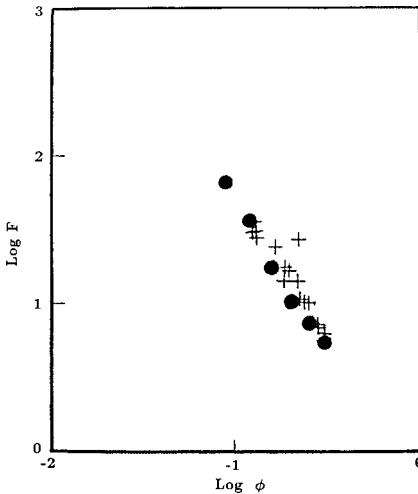


Figure 9.4 Comparison of the formation factor F of the grain consolidation model, computed by the random-walk method (circles) with the experimental data (+) (after Schwartz and Banavar, 1989).

geneous systems is rather old (Miller and Abrahams, 1960), Schwartz and Banavar (1989) appear to be the first to apply it to inhomogeneous porous media.

In contrast with diffusivity and conductivity, there is *no* random walk method for estimating the permeability of a porous medium because there is no *exact* relation between the effective permeability K_e and conductivity g_e , or between K_e and the effective diffusivity D_e . This is not totally surprising, as g_e and D_e are calculated from the solution of the Laplace's equation, which is a scalar equation, whereas K_e is calculated from the solution of the Stokes' equation, a vector equation. Thus, one cannot expect to have a general relation between the three quantities. This lack of a general relation between a scalar and a vector system is well-known in statistical physics.

Random walk concepts continue to play an important role in modeling of flow and transport in porous media. On the one hand, the technique has been extended for modeling of transport in fractured porous media (Pan and Bodvarsson, 2002). On the other hand, a particular type of random walks, namely, continuous-time random walks has been used successfully for modeling of dispersion and solute transport in porous media. The latter extension will be described in Chapters 11 and 12.

9.9.2

Lattice-Gas and Lattice-Boltzmann Methods

In this section, we describe two more recent methods for numerical simulation of flow in porous media (and fractured rock). The two approaches are the lattice-gas (LG) and lattice-Boltzmann⁶⁾ (LB) methods. The two methods may be used with practically any model of porous media.

9.9.2.1 Lattice-Gas Method

Broadwell (1964) was probably the first who developed a LG model for fluid flow problems. In his model, the fluid velocity was a discrete variable, but space and time were both continuous. Hardy, de Pazzis, and Pomeau (1973, 1976) (HPP) developed a completely discrete model for simulating fluid flow in the square lattice that could mimic several features of real flow problems. However, because a square network is not isotropic, the HPP model in the continuum limit did not reduce to the Navier–Stokes equation. As a result, the model did not receive much attention.

The LG method is based on large lattices in which each site can be in one of several discrete states. The lattice is populated by fictitious particles (representing the fluid), and the variables describing the state of the system are Boolean, indicating the presence or absence of the particles in the bonds of the lattice. The evolution of the system is governed by a set of collision rules that dictate how the particles move in the lattice and are scattered once they collide with each other (see below).

6) Ludwig-Eduard Boltzmann (1844–1906) was a Professor in universities in Vienna, Graz, Munich, and Leipzig. He developed kinetic theory of gases and the fundamental entropy-probability relation, $S = k_B \ln p$, which is engraved on his tombstone in Vienna (k_B is the Boltzmann constant).

Both time and space are discrete, and usually the connections between the sites are between nearest neighbors only – ideal conditions for high-speed simulations on vector or parallel computers.

The LG approximation of the Navier–Stokes equation in 2D is based on particles of unit mass either resting on the site of a lattice, or moving with unit velocity on one of the six bonds emanating from each lattice site. Frisch, Hasslacher, and Pomeau (1986) (FHP), using the HPP model, showed that in order for the discrete equations reduce to the usual Navier–Stokes equations, 2D simulations must be done with the triangular lattice (which is isotropic) on which the particles either have a unit velocity along a bond, or are at rest at a site. Up to six particles may reside at any site on the triangular lattice. Multiple occupancy of a site is, however, subject to the constraint that no two particles at a site can have the same velocity.

Figure 9.5 shows how the particles move on the triangular lattice. If they hit each other, they are scattered according to the laws of momentum conservation. For example, one particle hitting another one that is at rest may be scattered in such a way that its direction changes by 60° , and that the previously resting particle moves in a direction inclined at -60° with respect to the direction of the incoming particle. However, if only two-particle collisions are used, they would lead to anisotropy. One also needs three-particle collisions in order to remove the anisotropy (see Figure 9.5). Thus, two- and three-particle collisions constitute the minimum set of collision rules necessary to obtain isotropic behavior for the LG in the continuum limit. Usually up to four particle collisions are employed (five and six particle collisions almost never happen). If a particle hits a solid wall, it is reflected by 180° to simulate the no-slip boundary condition.

The extension of the LG to 3D networks is more complex since no regular 3D lattice is isotropic and, thus, in the continuum limit, one would have spurious terms in addition to those in the Navier–Stokes equation which are caused by the anisotropy of the lattice. There are, however, several ways of circumventing this difficulty. One can, for example, use a 3D topologically-random lattice, for example,

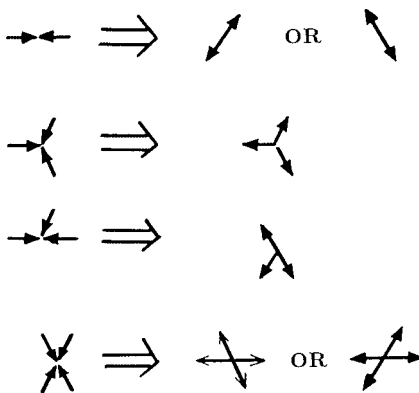


Figure 9.5 Collision rules for particle motion on a triangular lattice during lattice gas simulation of fluid flow.

the Voronoi lattice that is macroscopically isotropic (Sahimi, 1989). Alternatively, one can use (d'Humieres *et al.*, 1986) the FHP model on a *four-dimensional* face-centered hypercubic (FCHC) lattice. The nodes (x_1, x_2, x_3, x_4) of the lattice satisfy the condition that $x_1 + x_2 + x_3 + x_4$ is even, where the x_i are integer numbers. For this lattice, which has a coordination number of 24, all pairwise symmetric fourth-order tensors are isotropic and, thus, one can simulate the Navier–Stokes equation on such a lattice. We may then make the observation that any solution of four-dimensional equations that does not depend on the fourth dimension is a solution of the corresponding three-dimensional equations. This suggests the use of a FCHC lattice that wraps around periodically in the fourth direction.

In practice, a lattice is used that is only one lattice unit long in the fourth dimension and, therefore, has an effectively 3D structure. Figure 9.6 shows the 3D projection of an elementary cell of the FCHC lattice. The disadvantage of the model is that, although the fourth dimension is very thin, the discrete velocities still have components in *all* the directions. Therefore, the model is bit intensive (24 or 25 bits per site as compared with six in two dimensions).

A third approach (d'Humieres *et al.* (1986) is to use a 3D cubic lattice in which the particles move with speeds zero, one and $\sqrt{2}$ (instead of zero and one in 2D). The model uses only 19 bits per site. Note that having a large number of discrete particle velocities is not necessarily a disadvantage because it allows more collisions between the particles and, therefore, higher Reynolds number flows. Despite the discrete nature of the LG model, it is capable of exhibiting rich macroscopic complexity (d'Humieres and Lallemand, 1986).

To make the discussion more concrete, and also to lay the foundation for describing the LB, let us consider the LG on the triangular lattice and describe it in terms of the particle velocities. In this model, the particles have momenta selected from

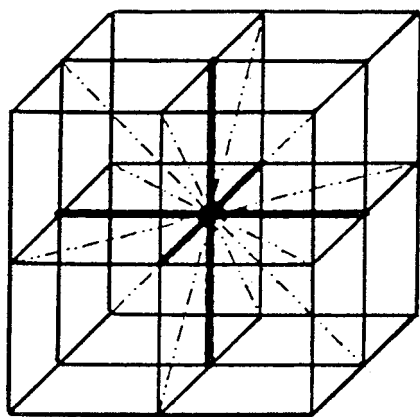


Figure 9.6 Three-dimensional projection of the elementary cell of the FCHC lattice. Two particles along the thick black lines can propagate with velocities $V = \pm 1$. The particle velocity along the dotted lines is zero.

the vector

$$\mathbf{u}_i = \left[\cos \frac{1}{3} \pi (i-1), \sin \frac{1}{3} \pi (i-1) \right], \quad i = 1, \dots, 6. \quad (9.135)$$

The microscopic densities corresponding to the number of particles and momentum conservation at position $\mathbf{x}$ at time t are, respectively

$$n(\mathbf{x}, t) = \sum_i n_i(\mathbf{x}, t) \quad (9.136)$$

and

$$\mathbf{u}_l(\mathbf{x}, t) = \sum_i \mathbf{u}_{i,l} n_i(\mathbf{x}, t), \quad (9.137)$$

where $n_i(\mathbf{x}, t)$ is the Boolean variable that indicates the presence ($n_i = 1$) or absence ($n_i = 0$) of a particle moving with momentum $\mathbf{u}_i$ at site $\mathbf{x}$ and time t . We have used $i, j, k, l, \dots$ to label the Cartesian coordinates. An equation for the evolution of the system can then be written down if we assume that the particles first move in the direction of their velocity vector and then collide with other particles. Under these conditions, the evolution of the system is described by

$$n_i(\mathbf{x} + \mathbf{v}_i, t + 1) = n_i(\mathbf{x}, t) + \Omega_i[n_i(\mathbf{x}, t)], \quad (9.138)$$

where Ω_i is the microscopic collision operator, which is the Boolean algebra expression that corresponds to the set of collisions described above, and $\mathbf{v}_i$ is the particle velocity vector.

To simulate flow through porous media with the LG model (as well as with the LB method), one first generates the configuration of the pore space. In their simulations with the LG, Rothman (1988) used rectangular obstacles and variations of them in his 2D simulations, while Brosa and Stauffer (1989, 1991), Duarte and Brosa (1990), Kohring (1991a,b,c), Sahimi and Stauffer (1991), and Knackstedt *et al.* (1993) used circular and overlapping or non-overlapping obstacles. The same simulations can be carried out with the LB method. One main advantage of using the LG or the LB is that *any* configuration of the pore space can be used. Thus, one may even utilize the digital image of a natural porous medium in the simulations, provided that a proper computational grid can be superimposed on the image. Once the desired pore space configuration is generated, the simulations can begin.

Let us emphasize once again that, in order for the LG method (as well as the LB method) to represent a physical flow phenomenon, (1) the lattice used in the simulation must have sufficient symmetry; (2) mass, momentum, and energy must be conserved, and (3) local equilibrium must exist and depend upon only the conserved quantities.

9.9.2.2 Lattice-Boltzmann Method

The LG method is not free of drawbacks. It suffers from, for example, statistical noise. This results in some physical phenomena that lead, among other things, to

the divergence of viscosity in 2D fluids. Moreover, the LG method has a velocity-dependent pressure which is unphysical. The applicability of the LG model is also limited to low flow velocities.

Various models have been proposed to circumvent the difficulties that the LG encounters (Rothman and Zaleski, 1997; Chen and Doolen, 1998; Sukop and Thorne, 2006). Several collision rules have also been proposed (Qian *et al.*, 1992; Rothman and Zaleski, 1997; Chen and Doolen, 1998). McNamara and Zanetti (1988), for example, proposed a LB model for simulating flow problems that retains the advantages that the LG method has, but does not have some of its drawbacks. In their method, $n_i(\mathbf{x}, t)$ in Eqs. (9.135) and (9.137) is a *continuous* variable between zero and one, and replaces the Boolean operations for the collisions with the appropriate arithmetic operations. Thus, instead of working with n_i , which is subject to large fluctuations and noise, one works with its mean value $N_i = \langle n_i \rangle$, which is no longer a Boolean variable. This implies that instead of following each particle in detail, one only considers the *average* behavior of a collection of them.

Such a scheme removes the statistical noise that the LG simulations usually suffer from. It can, however, be shown that this scheme is not practical for flow simulations in 3D systems because, in practice, the expression for Ω_i is a polynomial in N_i that has as many terms as there are collisions. As there can be up to 25 different collisions in 3D, the usefulness of the method is lost. Higuera *et al.* (1989) proposed a simpler method in which Ω_i is a linear function. Both methods, however, still contain a velocity-dependent pressure.

For concreteness, let us first describe one such set of collision rules that utilize nine velocities for the fluid particles in 2D systems. The pore space is discretized into a $L \times L$ lattice, where the lattice spacing is Δx . For each lattice point $\mathbf{x}$, we define at time t , a distribution function $n_i(\mathbf{x}, \mathbf{v}_i, t)$, where the velocity $\mathbf{v}_i$ can take on any of the nine values. The distribution function $n_i(\mathbf{x}, t) = n_i(\mathbf{x}, \mathbf{v}_i, t)$ satisfies the discrete Boltzmann equation:

$$n_i(\mathbf{x} + \mathbf{v}_i \Delta t, t + \Delta t) - n_i(\mathbf{x}, t) = -\frac{\Delta t}{\tau} [n_i(\mathbf{x}, t) - n_i^{\text{eq}}(\mathbf{x}, t)], \quad (9.139)$$

where Δt is the time step, τ is a relaxation time, and $n_i^{\text{eq}}(\mathbf{x}, t)$ is the quasi-equilibrium distribution function that, for low Mach numbers, is given by (Zau *et al.*, 1995; He and Luo, 1997; Madadi and Sahimi, 2003; Madadi *et al.*, 2003)

$$n_i^{\text{eq}}(\mathbf{x}, t) = \begin{cases} \frac{1}{9} \rho(\mathbf{x}, t) \left[1 + \frac{3}{v_i} \mathbf{v}_i \cdot \mathbf{u} + \frac{9}{2v_i^2} (\mathbf{v}_i \cdot \mathbf{u})^2 - \frac{3}{2} \mathbf{u}^2 \right], & i = 1, \dots, 4, \\ \frac{1}{36} \rho(\mathbf{x}, t) \left[1 + \frac{3}{v_i} \mathbf{v}_i \cdot \mathbf{u} + \frac{9}{2v_i^2} (\mathbf{v}_i \cdot \mathbf{u})^2 - \frac{3}{2} \mathbf{u}^2 \right], & i = 5, \dots, 8, \\ \frac{4}{9} \rho(\mathbf{x}, t) \left(1 - \frac{3}{2} \mathbf{u}^2 \right), & i = 9. \end{cases} \quad (9.140)$$

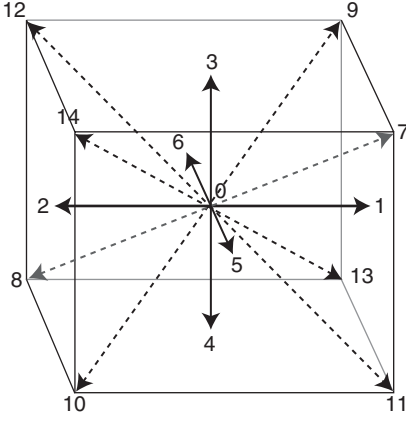


Figure 9.7 Possible directions for the motion of a particle at a lattice site in the d3q15 LB model. Solid arrows show the motion of class I particles with a speed of one, while dashed arrows indicate those of Class II particles with a speed of $\sqrt{3}$ (after Singh and Mohanty, 2000).

Here, $v_i = |\mathbf{v}_i|$, and $\rho(\mathbf{x}, t)$ and $\mathbf{u}(\mathbf{x}, t)$ are, respectively, the density and linear macroscopic momentum of the fluid given by

$$\rho(\mathbf{x}, t) = \sum_{i=1}^9 n_i^{\text{eq}}(\mathbf{x}, t) = \sum_{i=1}^9 n_i(\mathbf{x}, t), \quad (9.141)$$

$$\rho(\mathbf{x}, t) \mathbf{u}(\mathbf{x}, t) = \sum_{i=1}^9 \mathbf{v}_i n_i^{\text{eq}}(\mathbf{x}, t) = \sum_{i=1}^9 \mathbf{v}_i n_i(\mathbf{x}, t). \quad (9.142)$$

One can show using a Chapman–Enskog expansion and Eq. (9.140) that the continuity and Navier–Stokes equations are recovered at large times and length scales. We define the lattice spacing Δx as the unit of length and the time step Δt as the unit of time. The viscosity μ of the fluid is then given by $\mu = (1/6)(2\tau - 1)\rho$, and the pressure by $P = c_s \rho$, with $c_s = 1/3$.

As in 2D, an appropriate choice of a 3D lattice guarantees isotropy for the macroscopic equations in the continuum limit. Two types of lattice that are commonly used are the d3q15 and d3q18 models. Here, d3 indicates three dimensions, and q15 and q18 indicates the number of links (Maier *et al.*, 1996). As in the LG and 2D LB methods, the particles could be at rest. The d3q15 lattice does include such particles, but the d3q18 model does not. For example, in Figure 9.7, the indices 0–14 show the possible directions of motion available for a particle in the d3q15 model. Six lattice vectors, labeled 1–6 and referred to as Class I, point to the six nearest-neighbor sites. Eight lattice vectors, labeled 7–14 and referred to Class II, point along the body diagonals to the next sites along the diagonals. The zero at the center of Figure 9.7 indicates the stationary particles.

To simplify the notations that we used for the 2D model, we assume a unit time step for a displacement $\mathbf{d}_i$. Then, the discrete Boltzmann equation is given by an

equation similar to Eq. (9.138):

$$n_i(\mathbf{x} + \mathbf{d}_i, t + 1) = n_i(\mathbf{x}, t) + \Omega_i(\mathbf{x}, t), \quad i = 1, \dots, q. \quad (9.143)$$

Here, Ω_i is the collision operator that drives each particle population toward its equilibrium. It is given by the Bhatnagar–Gross–Krook approximation (Bhatnagar *et al.*, 1954),

$$\Omega_i = -\tau^{-1}(n_i - n_i^{\text{eq}}). \quad (9.144)$$

As in the 2D case, the equilibrium distribution should be such that the formulation reduces to the low-Mach number limit of the Navier–Stokes equations with *Galilean invariance* (i.e., the principle that the laws of physics are the same in all inertial frames). For the d3q15 model, in which the fluid particles' velocities can be zero or one, or $\sqrt{3}$, one has

$$n_i^{\text{eq}} = \begin{cases} \rho \left[\frac{1}{8} - \frac{1}{3} u^2 \right], & \text{stationary particles} \\ \rho \left[\frac{1}{8} - \frac{1}{3} (\mathbf{d}_i \cdot \mathbf{u}) + \frac{1}{2} (\mathbf{d}_i \cdot \mathbf{u})^2 - \frac{1}{6} u^2 \right], & \text{Class I particles,} \\ \rho \left[\frac{1}{64} + \frac{1}{24} (\mathbf{d}_i \cdot \mathbf{u}) + \frac{1}{16} (\mathbf{d}_i \cdot \mathbf{u})^2 - \frac{1}{48} u^2 \right] & \text{Class II particles.} \end{cases} \quad (9.145)$$

The equation of state relating the pressure to the fluid density is given by $P = c_s^2 \rho$, where $c_s = \sqrt{3/8}$ for the d3q15 model. The expression for the viscosity is the same as in 2D. If a body force $\mathbf{f}$, such as gravity, also acts on the fluid, then the streaming operation is supplemented with a forcing function at each node of the lattice,

$$n_i(\mathbf{x}, t) \rightarrow n_i(\mathbf{x}, t) + \mathbf{d}_i \mathbf{f}, \quad i = 1, \dots, q. \quad (9.146)$$

On the internal surface of the pores, the flow field must satisfy the no-slip boundary condition. The simplest way of implementing this condition is the bounce-back rule in which a particle incident on the surface reverses its direction. Thus, if, for example, the walls are stationary, the bounce-back rule implies that the unknown populations of the inward-pointing links are assigned the known values of their outward-pointing counterparts on the solid walls. The bounce-back rule is straightforward to implement and is applicable to all complex solid surface geometries, but its accuracy could be limited. In fact, it has been shown to be accurate to first order in space. Inamuro *et al.* (1995) showed that the bounce-back rule becomes inaccurate if $\tau/\Delta t \geq 3$. Other rules for implementing the no-slip boundary condition that may be more accurate have also been suggested (Ginzbourg and Adler, 1994; Inamuro *et al.*, 1995), but they are also more complex than the bounce-back rule to implement, particularly on rough surfaces. A good discussion of implementing the no-slip boundary conditions is given by Singh and Mohanty (2000).

To begin the simulations, the boundary conditions must be specified, and the initial distribution $n_i(\mathbf{x}, 0)$ for the time $t = 0$ must be supplied. One can, for example, apply a constant pressure to the porous medium at its upstream face, and the fluid velocities at the entrance of all the pores that are directly connected to this side are also assumed to be constant over their cross sections. As for $n_i(\mathbf{x}, 0)$, one may use, for example, the equilibrium distribution n_i^{eq} . In fact, if steady-state flow is of prime interest, any initial distribution or condition may be used (Skordos, 1993). The system then evolves according to Eqs. (9.140)–(9.142), or (9.147). Since the pressure is given by $P = c_s \rho$, it might appear that the fluid is compressible. However, since the Mach number is small, the fluid is only very weakly compressible.

We should point out that the method can also be used for simulating flow through a fracture network (Madadi and Sahimi, 2003, 2011). All one must do is generate the fracture network configuration, after which, simulation of flow through the network is similar to that through porous media. We will come back to this point in Chapter 12.

It should also be pointed out that due to the ease with which flow in complex geometries of a pore space can be simulated with the LB method, one can use it to estimate the permeability of a 3D porous medium with a given geometry and compare the results with various rigorous results and bounds for the permeability of the same system described earlier in this chapter. Cancelliere *et al.* (1990), for example, used a LB method to undertake such a study for the Swiss-cheese or randomly-distributed penetrable sphere model described in Chapters 3 and 6. They found that at high solid fractions the calculated permeability is within one order of magnitude of the upper bound due to Weissberg and Prager (1970) and Berryman and Milton (1985).

The LB has become a standard tool for simulating not only single- and multiphase flow in porous media, but also for studying a wide variety of problems involving multicomponent fluids, phase transitions, and so on. Illuminating applications of the LB method to porous media problems were reported by Singh and Mohanty (2000) and Hidajat *et al.* (2002).

9.10

Relation between Permeability and Electrical Conductivity

An exact and general relation between the effective permeability and electrical conductivity of porous media does not exist. Although many empirical and semi-empirical relations between K_e and g_e have been proposed in the past, almost all of them are accurate only for certain classes of porous media, but not for all porous media. For example, although Wong *et al.* (1984) found that $g_e \sim \phi^m$, and $K_e \sim \phi^{m'}$ with $m' = 2m$, the relation is restricted to their particular model and is not expected to hold for a general porous medium.

Johnson *et al.* (1986), however, introduced a well-defined parameter $\mathcal{A}$, defined by

$$\mathcal{A} = 2 \frac{\int |E(\mathbf{r})|^2 dV_p}{\int |E(\mathbf{r})|^2 dS_p} \neq \frac{V_p}{S_p}, \quad (9.147)$$

where $E(\mathbf{r})$ is the potential in the electrical conduction problem in a porous medium that is saturated with a conducting fluid (such as brine), and V_p and S_p are, respectively, the pore volume and the pore surface area. Note that V_p/S_p is a *geometrical* parameter that can be measured and is *independent* of any flow or transport process. On the other hand, $\mathcal{A}$ is a *dynamical property*, defined for the specific problem of electrical conduction and, therefore, cannot be measured by geometrical analysis alone. Since $E(\mathbf{r})$ vanishes in certain regions of the pore space (for example, in the isolated and dead-end region), $\mathcal{A}$ is roughly a measure of the *dynamically connected* pores of the medium, that is, those that belong to the backbone of the flow or transport path. Johnson *et al.* (1986) proposed that for 3D porous media,

$$K_e \simeq c \frac{\mathcal{A}^2}{8F} = c \frac{\mathcal{A}^2}{8} \frac{g_e}{g_f}, \quad (9.148)$$

where $c \sim \mathcal{O}(1)$, and $F = g_f/g_e$ is the formation factor, with g_f being the conductivity of the fluid. It should be pointed out that, for the obvious reasons discussed earlier, Eq. (9.148) cannot *generally* be exact, although due to the physical meaning of $\mathcal{A}$, Eq. (9.148) represents an appealing relation between K_e and g_e . Various groups tested the validity of Eq. (9.148) (Banavar and Johnson, 1987; Straley *et al.*, 1987; Banavar *et al.*, 1988; Schwartz and Banavar, 1989; Saeger *et al.*, 1991; Kostek *et al.*, 1992; Schwartz *et al.*, 1993) using a variety of numerical and analytical methods as well as experimental data for well-defined porous media. Avellaneda and Torquato (1991) investigated the relation between K_e and g_e , and derived the conditions under which an approximate relation between the two may be expected. It appears that (Kostek *et al.*, 1992), unless a porous medium contains two widely different relevant length scales, Eq. (9.148) should be very accurate.

9.11

Relation between Permeability and Nuclear Magnetic Resonance

Timur (1968, 1969a,b) suggested that nuclear magnetic resonance (NMR) may be used as a technique for measuring the effective permeability K_e of a porous medium. At first, this may seem to be impractical because most of the fluid in a porous medium is stored in the pore bodies, whereas K_e and the transport properties are controlled by the size of the narrow pore throats. Indeed, as described in Chapter 4, the NMR has been used to measure fluid saturations in multiphase flows in porous media (Mandava *et al.*, 1990; Chen *et al.*, 1994; Liaw *et al.*, 1996), which are directly related to the spatial distribution of the porosity.

The possibility of using the NMR for measuring the effective permeability was studied (for example, de Gennes, 1982 and Banavar *et al.*, 1985) in the 1980s. In

particular, Kenyon *et al.* (1988) studied the relation between the NMR and the effective permeability of 56 water-saturated sandstones. They found that the decay of proton magnetization $M_z(t)$ is described by a stretched exponential

$$M_z(t) = m_0 \exp \left[- \left(\frac{t}{T_1} \right)^{\delta_1} \right], \quad (9.149)$$

where T_1 is a relaxation time described below. In an early work, Seevers (1966) estimated the effective permeability of a uniform sandpack using the relation, $K_e \sim \phi T_1^2$, where ϕ is the porosity. Kenyon *et al.* (1988) found, however, that K_e exhibits a very strong correlation with $\phi^4 T_1^2$. Figure 9.8 presents their data plotted in their proposed fashion, from which one obtains

$$K_e \sim (\phi^4 T_1^2)^{\delta_2}, \quad (9.150)$$

with $\delta_2 \simeq 0.7$. Billardo *et al.* (1991) carried out NMR experiments on, and measured the permeabilities of, 44 sandstones and found that Eq. (9.150) fits their data very well. Banavar and Schwartz (1987) investigated the same problem in the grain-consolidation model of Roberts and Schwartz (1985) and reached the same conclusion.

These results may seem surprising, that is, until we ask ourselves: what is the physical significance of the relaxation time T_1 ? The protons in the hydrogen of water molecules carry a nuclear magnetic moment which enables them to align themselves with an externally applied field. However, because water molecules at

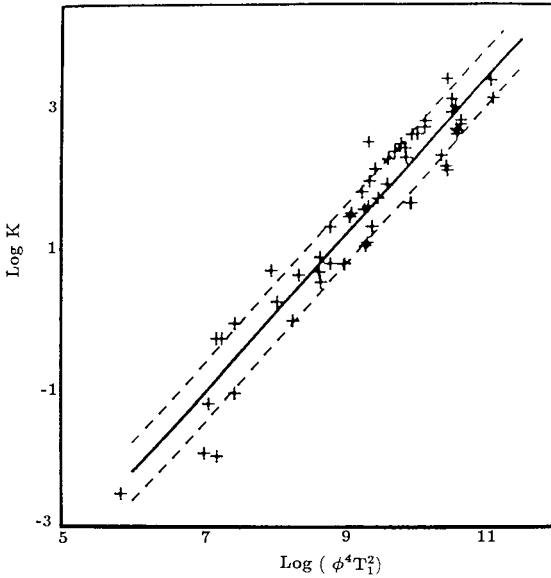


Figure 9.8 Correlation of the effective permeability K_e of water-saturated sandstones with the porosity ϕ and the NMR relaxation time T_1 (after Kenyon *et al.*, 1988).

room temperature are thermally agitated, only a few of the protons actually align themselves with the external field that are, however, detectable. If the external field is removed, the porous medium and its content will go back to the equilibrium state. The time for reaching the equilibrium is also the same as the time that the system requires for building up its external magnetization after the external field has been applied. This time is usually denoted by T_1 , and is called the *spin-lattice relaxation time* for protons.

Why should T_1 be related to K_e ? Experiments indicate that if one measures T_1 for water saturating a porous medium, one finds T_1 to be much smaller than the corresponding time for the same water in the bulk. This is because T_1 is affected strongly by pore surface relaxation mechanisms and, thus, it is expected to be sensitive to the microstructure of the porous medium. This sensitivity explains why T_1 may provide insight into and information about the structure of the pore space.

Thompson *et al.* (1989) used deuterium NMR to study sandstones, carbonate rock, and synthetic porous samples. The reason for using deuterium, instead of proton, is that (Williams and Fung, 1982) it has a much smaller magnetic moment than protons. They measured T_{1p} , the longitudinal relaxation time in the rotating frame, and found that the magnetization $M_z(t)$ follows a stretched-exponential similar to Eq. (9.149), with T_1 replaced by τ_{SE} , the corresponding stretched-exponential relaxation time. Thompson *et al.* argued that the length scale ℓ_c , which they introduced (see Chapter 10) for estimating the effective permeability of a porous medium, is proportional to a time scale τ_{NMR} , the NMR relaxation time. Their experimental data appeared to be in complete agreement with the argument. In fact, Thompson *et al.* showed that the data were fully consistent with

$$K_e \sim (T_{1p} \phi)^2 \sim (\tau_{NMR} \phi)^2, \quad (9.151)$$

which is consistent with Eq. (9.150), except that the exponents of T_{1p} (or τ_{NMR}) and T_1 in the two equations are not the same. However, Thompson *et al.* (1989) also pointed out that their data could also be fitted with an equation similar to Eq. (9.150), but that the quality of the resulting fit would be slightly worse than that provided by Eq. (9.151). Thompson *et al.* (1989) also showed that T_{1p} is proportional to the *width* of the pore size distribution of the porous medium as well as the water saturation in a partially-saturated porous medium.

Others (LaTorraca *et al.*, 1993; Kubica, 1995; Straley *et al.*, 1995) suggested that a more accurate correlation exists between the effective permeability of sandstones and the logarithmic mean of the relaxation time (since in a disordered porous medium, there is a distribution of such relaxation times). Hidajat *et al.* (2002) generated by computer simulation spatially-correlated models of porous media, and computed their effective permeability and the NRM relaxation responses. Their computations indicated that in order to have an accurate correlation between K_e and the NMR relaxation time, three conditions must be met. (1) The surface relaxivity should not vary very significantly. (2) The formation factor F should depend only on the porosity, and (3) the characteristic pore body radius should be proportional to the characteristic pore throat radius.

Since the NMR measurements can be made at depth in the earth with well-logging tools, they provide one of the few in-situ techniques of estimating the effective permeabilities (and saturations). Indeed, the technique is now used in practice (Mair *et al.*, 1999; Wang *et al.*, 2004, 2005; Glover *et al.*, 2006).

9.12

Dynamic Permeability

So far, we have described the properties of the static permeability of a porous medium. However, what can one say about the dynamic (frequency-dependent) permeability $\tilde{K}_e(\omega)$? If one measures the dynamic permeability, can the data provide *extra* information in addition to what the static permeability $\tilde{K}_e(\omega = 0) = K_e = K_0$ provides about the morphology of the porous medium? The dynamic permeability is defined by the generalized Darcy's law

$$\langle \mathbf{v}(\omega) \rangle = -\frac{\tilde{K}_e(\omega)}{\mu} \nabla P(\omega), \quad (9.152)$$

where ω is the frequency, and $\nabla P(\omega) = \nabla P \exp(-i\omega t)$. The properties of the dynamic permeability were studied by several groups in the 1980s and early 1990s. Auriault *et al.* (1985) carried out measurements of $\tilde{K}_e(\omega)$ to test the validity of Eq. (9.152). Charlaix *et al.* (1988b) measured $\tilde{K}_e(\omega)$ for a variety of fused-glass beads and crushed glass in the frequency range 0.1–1 kHz. Sheng and Zhou (1988), Zhou and Sheng (1989), Johnson *et al.* (1987), Chapman and Higdon (1992), and Knackstedt *et al.* (1993) studied the problem theoretically and by computer simulations. (Some errors in the studies of Sheng and Zhou were corrected by Chapman and Higdon, 1992). Let us summarize the most important results of such studies.

The starting point is Eq. (9.152) together with

$$\tilde{\tau}(\omega)\rho \frac{\partial \langle \mathbf{v} \rangle}{\partial t} = -\nabla P, \quad (9.153)$$

where $\tilde{\tau}(\omega)$ is the frequency-dependent tortuosity (a dimensionless quantity), and ρ is the density of the fluid. $\tilde{\tau}(\omega)$ and $\tilde{K}_e(\omega)$ are related:

$$\tilde{K}_e(\omega) = \frac{i\mu\phi}{\tilde{\tau}(\omega)\omega\rho}, \quad (9.154)$$

where $i = \sqrt{-1}$. As $\omega \rightarrow 0$, we should have $\tilde{K}_e(\omega) \rightarrow K_0$, where K_0 is the static permeability and

$$\lim_{\omega \rightarrow 0} \tilde{\tau}(\omega) = \frac{i\mu\phi}{K_0\omega\rho}. \quad (9.155)$$

Johnson *et al.* (1987) showed that in the high-frequency limit,

$$\lim_{\omega \rightarrow \infty} \tilde{\tau}(\omega) = \tau_\infty + \frac{C_\infty}{\sqrt{-i\omega}}, \quad (9.156)$$

where $C_\infty = K_0 K_B / (\mu \phi)$, with K_B being the bulk modulus of the fluid. Moreover, one has

$$\tau_\infty = \phi F, \quad (9.157)$$

for any porous medium, a result that is generally attributed to Lord Rayleigh.⁷ Johnson *et al.* (1987) also showed that in the high-frequency limit,

$$\lim_{\omega \rightarrow \infty} \tilde{\tau}(\omega) = \tau_\infty \left[1 + \sqrt{\frac{i\mu}{\omega\rho} \frac{2}{\Lambda}} \right], \quad (9.158)$$

$$\lim_{\omega \rightarrow \infty} \tilde{K}_e(\omega) = \frac{i\mu\phi}{\tau_\infty\omega\rho} \left[1 - \sqrt{\frac{i\mu}{\omega\rho} \frac{2}{\Lambda}} \right], \quad (9.159)$$

so that the *high-frequency* limit provides information on Λ and, thus on K_0 . Moreover, Johnson *et al.* (1987) proposed that

$$\tilde{\tau}(\omega) = \tau_\infty + \frac{i\mu\phi}{\omega K_0\rho} \sqrt{1 - \frac{4i\tau_\infty^2 K_0^2 \omega\rho}{\mu \Lambda^2 \phi^2}}, \quad (9.160)$$

$$\tilde{K}_e(\omega) = K_0 \left(\sqrt{1 - \frac{4i\tau_\infty^2 K_0^2 \omega\rho}{\mu \Lambda^2 \phi^2}} - \frac{i\tau_\infty K_0 \omega\rho}{\mu \phi} \right)^{-1}. \quad (9.161)$$

These results, together with those of Sheng and Zhou and the experimental data of Charlaix *et al.* (1988b), also indicate that $\tilde{K}_e(\omega)$ generally follows a scaling equation,

$$\frac{\tilde{K}_e(\omega)}{K_0} = f\left(\frac{\omega}{\omega_c}\right), \quad (9.162)$$

where $\omega_c = \mu\phi/(\tau_\infty K_0\rho)$ is a characteristic frequency at which a crossover in $\tilde{K}_e(\omega)$ from a viscous flow regime to an inertial one takes place. The function $f(\omega/\omega_c)$ was found to be *universal*, independent of the microstructure of a porous medium.

Johnson (1989) showed that the above predictions, when compared with the results of numerical simulations, are very accurate. Therefore, although Eq. (9.162) implies that one cannot gain much microstructural information about the porous medium by simply measuring $\tilde{K}_e(\omega)$ (as $f(\omega/\omega_c)$ is universal), Eq. (9.158) may still be used to estimate $K_0 = K_e$, the static permeability of the porous medium (via its relation with Λ). The experimental data of Charlaix *et al.* (1988b) as well as the analytical and numerical calculations of Sheng and Zhou (1988), Chapman and Higdon (1992) and Knackstedt *et al.* (1993) all supported the validity of Eq. (9.162). Cortis *et al.* (2003) studied the influence of pore roughness on high-frequency permeability.

7) John William Strutt, 3rd Baron Rayleigh (1842–1919), universally known as Lord Rayleigh, was an English physicist and co-discoverer (with William Ramsay) of the element argon, for which he was awarded the Nobel Prize for Physics in 1904. He made tremendous contributions to science including, among others, Rayleigh scattering, Rayleigh (surface) waves, and theory of sound.

9.13

Non-Darcy Flow

As in any fluid flow problem, the range of validity of Darcy's law is expressed in terms of a Reynolds number Re defined in terms of a characteristic length of the system in which fluid flow occurs. For flow through unconsolidated porous media, however, it is customary to express the Re in terms of a characteristic grain size, and for flow in consolidated porous media in terms of a mean pore size. It has been suggested by some to use $\sqrt{K_e/\phi}$ as the characteristic length, as K_e has the units (length)², while others have used $\sqrt{K_e}$. In any event, it is generally believed that Darcy's law is applicable as long as $Re \leq 10$. This is the flow regime in which viscous forces are predominant. Beyond $Re = 10$, there is a transition zone in which inertial forces begin to be important and eventually predominant. Roughly speaking, for $Re > 100$, the transition zone ends and turbulent flow regime begins.

It was pointed out in Section 9.3 that Darcy's law may also break down for flow of gases at low pressures through porous media, and since many gases (such as air) are used for measuring the effective permeability of porous media, the deviations from Darcy's law may be important. For example, if a pore diameter is comparable with the mean-free path of a molecule, that is, the mean distance that it travels before it collides with another molecule, then deviations from Darcy's law become significant. This breakdown may be due to the fact that at low pressures, the no-slip boundary condition on the pore surface is no longer valid. Gas molecules diffuse and flow on the pores surface and contribute to the overall flux. This phenomenon is usually called the Klinkenberg effect, and represents the well-known Knudsen flow.

If the fluid velocity is high enough that Darcy's law is not valid, then one must add some correction terms to the basic Darcy's law in order to take into account the effect of inertia. The first attempt in this direction was probably made by Forchheimer (1901), who suggested that for a pressure gradient $\partial P/\partial x$, one must write

$$-\frac{\partial P}{\partial x} = \frac{\mu}{K_e} \cdot V + \rho\beta V^2, \quad (9.163)$$

where V is the magnitude of the fluid velocity vector, β is a constant, and x is the direction of macroscopic flow. Note that the Forchheimer equation is equivalent to the Ergun equation, Eq. (9.102). Many theoretical attempts have been made to understand the effect of inertia on flow through porous media, and to derive Forchheimer equation (Joseph *et al.*, 1982; Ruth and Ma, 1988; Mei and Auriault, 1991; Ma and Ruth, 1993; Whitaker, 1996).

The dependence of the parameter β on the porous media characteristics has been studied extensively. From the equivalence of Forchheimer and Ergun equations, we obtain

$$\beta = 10^4 \frac{a}{\sqrt{b}} K_e^{-\frac{1}{2}} \phi^{-\frac{3}{2}}, \quad (9.164)$$

with $a = 1.75$ and $b = 150$. From the equation suggested by MacDonald *et al.* (1979), Eq. (9.103), one obtains $b = 180$, and $1.8 \leq a \leq 4$. Much earlier, Janicek

and Katz (1955) had suggested that

$$\beta = 1.82 \times 10^8 K_e^{-\frac{5}{4}} \phi^{-\frac{3}{4}} . \quad (9.165)$$

Based on extensive data for both consolidated and unconsolidated porous media, Geertsma (1974) proposed that

$$\beta = 5.6 \times 10^{-3} K_e^{-\frac{1}{2}} \phi^{-\frac{11}{2}} , \quad (9.166)$$

while Liu *et al.* (1995) proposed that

$$\beta = 2.94 \times 10^7 (K_e \phi)^{-1} \tau , \quad (9.167)$$

where τ is the tortuosity factor of the porous medium. Huang and Ayoub (2008) discussed the applicability of the Forchheimer equation for describing non-darcy flows in oil reservoirs.

Andrade *et al.* (1999) studied the effect of inertia on fluid flow through a model porous medium, in which square obstacles are removed at random from a square network. They identified a transition from linear (Darcy-type) flow to a nonlinear one. Hill *et al.* (2001) used the lattice-Boltzmann method to examine the effect of inertia at small Reynolds numbers on flow through simple cubic, face-centered cubic, and body-centered cubic as well as random arrays of spheres. The solid volume fraction studied was anywhere from very small to the close-packed limit. They showed that the first inertial contribution to the drag force (and, hence, to the permeability) when scaled with the Stokes drag force on a single sphere in an unbounded flow is proportional to the square of the Reynolds number, and that the scaling holds, in fact, all the way to the close-packed limit, and that the first inertial contribution to the drag force relative to that of Stokes flow *decreases* with increasing the solid volume fraction.

To obtain a better fit of the experimental data, a third-order term has also been proposed:

$$\frac{\Delta P}{L} = a_1 V + a_2 V^2 + a_3 V^3 , \quad (9.168)$$

where a_1 , a_2 , and a_3 are fitted to the data, and are presumably dependent on the porosity and other characteristics of the porous medium.

10

Single-Phase Flow and Transport in Porous Media: The Pore Network Approach

Introduction

In Chapter 9, we studied the phenomena of single-phase flow and transport in porous media based on what we refer to as the continuum models. In the present chapter, we study the same in pore network models of porous media, using a variety of analytical and numerical techniques. As in Chapter 9, we mainly focus on low Reynolds-number flow, where Darcy's law is applicable. Experience with homogeneous materials – porous or not – as well as the general theory of flow and transport processes indicate that any *small* departure from equilibrium, which is represented by the existence of a gradient of intensity ∇G in the materials, gives rise to an associated flux J given by a linear relation

$$J = -\mathcal{T} \cdot \nabla G, \quad (10.1)$$

where the proportionality factor $\mathcal{T}$ is a *local* transport coefficient that characterizes the material at the *local* level. Equation (10.1) is also consistent with Darcy's law, for which $G = P$, and $\mathcal{T} = K/\mu$. In general, $\mathcal{T}$ is a second-rank tensor when a scalar quantity is transported or convected in the medium, and a fourth-rank tensor when ∇G represents the gradient of a vector quantity, such as displacement. If the material's morphology is isotropic, the resulting symmetry conditions on $\mathcal{T}$ simplify its form greatly, in which case a single transport coefficient may suffice for the description of, say, thermal conductivity, just as in many cases a single permeability suffices for describing fluid flow in a porous medium. As the discussion in Section 9.1 demonstrated, if the material is statistically homogeneous (but microscopically disordered), we expect a linear relation such as Eq. (10.1) to also exist between an imposed *small* average gradient of intensity $\langle \nabla G \rangle$ and the resulting *average* flux $\langle J \rangle$:

$$\langle J \rangle = -\mathcal{T}_e \cdot \langle \nabla G \rangle, \quad (10.2)$$

where $\mathcal{T}_e$ is now the effective transport coefficient that characterizes the material *macroscopically*, and the averages are over the ensemble of all possible configurations of the disordered material.

As the description of the continuum models and their flow and transport properties in Chapter 9 demonstrated, $\mathcal{T}_e$ differs, in general, from a simple average of the statistical distribution of the local flow or transport coefficient $\mathcal{T}$ because the microstructure of a porous material controls the effective flow and transport properties in a nontrivial manner. If detailed information is available on the materials' morphology, then it is exploited to the full for estimating their flow and transport properties, which is what we try to accomplish throughout this book. If, on the other hand, little is known about the morphology, one would be forced to fall back on observations, experience or hypothesis, or purely phenomenological theories that are, in essence, *speculations*.

Restricting the study to linear flow and transport properties of heterogeneous porous materials, we discover an important fact: a remarkably large number of seemingly distinct flow and transport problems can be analyzed by a common theoretical framework. For example, (1) thermal conduction; (2) electrical conduction; (3) diffusion, and (4) fluid flow are all described by Eq. (10.2), where the coefficient $\mathcal{T}_e$ is called, respectively, (1) thermal conductivity; (2) electrical conductivity; (3) diffusion coefficient, and (4) the ratio K_e/μ , where μ is the fluid viscosity. At steady state, the flux J in all the four phenomena satisfies a conservation equation:

$$\nabla \cdot J = 0. \quad (10.3)$$

Table 10.1 lists several flow and transport phenomena that are mathematically equivalent. It is precisely their mathematical equivalency that makes it possible to analyze these phenomena within a common theoretical framework.

Due to the analogy between conduction, diffusion, and fluid flow, one may develop a unified approach to all three phenomena based on the pore network models. We describe the approach in this chapter.

Table 10.1 Mathematically equivalent flow and transport problems with the corresponding flux J and flow or transport coefficient $\mathcal{T}$.

Phenomenon	Potential	Driving force	J	$\mathcal{T}$
Heat conduction	Temperature	Temperature gradient	Heat flux	k
Electrical conduction	Electric potential	Electric field	Current density	g
Diffusion	Density	Density gradient	Diffusant current density	$\mathcal{D}$
Fluid flow	Pressure	Pressure gradient	Fluid flux	K/μ

10.1

The Pore Network Models

The idea that a porous medium can be mapped onto an equivalent network of interconnected pore throats and pore bodies is rather old, and goes back to the 1950s. Mohanty (1981) and Lin and Cohen (1982) endowed the idea with a solid theoretical foundation. In particular, as described in Chapter 3, Mohanty (1981) described a systematic procedure by which the mapping should be carried out. Because most porous media are disordered, the mapping involves judicious choices of what constitutes a pore throat or pore body, and where they are connected. The network model can then be used for not only estimating the permeability of porous media, but also the effective diffusivity of gases and liquids in their pore space, and the effective conductivity or resistivity of the pore space if it is saturated by a conducting fluid (such as brine). The exact mapping of a porous medium onto an equivalent pore network is difficult. The macroscopic properties may also not be sensitive to some (but not all) microstructural details of the porous medium. Thus, in practice, a pore network is developed in which important structural details, such the average connectivity (coordination number), and pore size and pore length distributions are included. Chapter 4 already described the techniques for measuring the three characteristics.

Once the three characteristics are measured, the next step in the development of a pore network model is selecting a shape for the pore throats, represented by the network's bonds. They may, for example, be cylindrical or slit-like, or have converging-diverging segments. The shape of the cross-sectional area of the pore throats is not so important for determining single-phase flow and transport properties, but the same is not true for multiphase flows that will be studied in Chapters 14 and 15. If the shape of the pore throats is simple enough, then the problem of creeping (Darcy) flow in them can be solved analytically, which simplifies the computations with the pore network significantly. However, the availability of the analytical solution is not a prerequisite for developing a pore network model. One may utilize full numerical simulations in order to compute the flow field within the pore throats as well. The pore bodies are typically assumed to be spherical and large enough that the pressure drop in them can be neglected, so that each pore body (each node of the network) may be represented by a single (averaged) pressure. Other pore body shapes may also be used if one is willing to carry out extensive numerical simulations.

From the analytical solution of the fluid flow problem in a single pore throat, one derives an expression for the volume flow rate Q_{ij} in a pore throat $i - j$ in terms of the pressure drop along the pore throat, its length and effective radius, and the fluid viscosity. For example, for slow flow in a cylindrical pore throat of radius R_{ij} and length l_{ij} , one has

$$Q_{ij} = \frac{\pi R_{ij}^4 (P_i - P_j)}{8\mu l_{ij}}, \quad (10.4)$$

where both R_{ij} and l_{ij} follow their own statistical distributions that can, in principle, be measured. One then writes down a mass balance for each node (pore body) i , which mean that the *net* mass flow rate reaching the node is zero. Therefore,

$$\sum_{j \in \{i\}} \rho Q_{ij} = 0, \quad (10.5)$$

where ρ is the fluid density, and the sum is over the set of pore bodies (nodes) j that are connected to i by a pore throat. If the fluid is incompressible, then the fluid density ρ cancels out. In case of a compressible fluid, for example, a gas, ρ is expressed by an equation of state in terms of the pressure, in which case the problem may become nonlinear. In this chapter, we assume that the fluid is incompressible.

Writing down Eq. (10.5) for every interior node of the network results in a set of simultaneous equations for the nodal pressures. The boundary conditions must then be specified. For example, one may specify the pressures at two opposite faces of the network, and assume impermeable boundaries in the other direction(s), or utilize any other boundary conditions. The set of the equations subject to the given boundary conditions are then solved numerically. The most efficient numerical method for solving the set of the equations is the conjugate-gradient method. Once the pressure field throughout the network is determined, the effective permeability of the pore network is computed easily by calculating the total volume flow rate, and hence the mean flow velocity, through the network and utilizing Darcy's law.

Since the pore network can never be as large as the actual porous medium, one must utilize a Monte Carlo method in order to obtain accurate estimates of the properties of interest. This means that after computing the effective permeability of the pore network by the aforementioned procedure, one redistributes the pore sizes and lengths – that is, generates another *realization* of the pore network – repeats the above procedure and computes another estimate of the effective permeability. The procedure is repeated many times until the average effective permeability, with the averaging taken over the ensemble of the realizations, no longer changes if more realizations are made and the ensemble is expanded. In addition, unless the scale-dependence of the effective permeability is of interest, one must select an overall size (length) of the network such that its effective permeability would not change significantly if larger networks are used.

Various versions of such a basic pore network models have been used over the last six decades. As already mentioned in Chapters 4 and 5, Owen (1952) appears to be the first who carried out computations with a three-dimensional (3D) network of large pore bodies and very narrow pore throats in order to estimate the resistivity of sample porous media. Fatt (1956a,b,c) used a 2D network of pore throats with distributed sizes, and calculated the permeability and relative permeabilities for two immiscible fluids that will be studied in Chapters 14 and 15. In the 1960s, there were several works in which pore network models were used for calculating the permeability, conductivity and the formation factors, and investigating the relationship between them (Fatt, 1960; Nicholson, 1968; Rink and Schopper, 1968; Greenberg and Brace, 1969; Weinbrandt and Fatt, 1969). Use of the pore network model was continued in the 1970s (Nicholson and Petropoulos, 1971, 1975, 1977; Shankland

and Waff, 1974; Dullien, 1975), 1980s (Koplik, 1982; Koplik *et al.*, 1984; Seeburger and Nur, 1984; Doyen, 1988; Constantinides and Payatakes, 1989), 1990s (David *et al.*, 1990) and beyond (Mukhopadhyay and Sahimi, 2000). These works vary in the amount of the microstructural details that were included in the pore network model, the sizes of the networks that were used, and so on, though the essential idea used in all of them is the same as that described.

On the other hand, Bryant *et al.* (1993a,b) developed a pore network model based on Finney's random close packing of equal spheres (Finney, 1970), *without* invoking any major assumption regarding the microstructure of the network, and calculated the permeability of the network. Øren and Bakke (2002) used a similar method to study flow problems, while Dadvar and Sahimi (2003) used the technique in order to study flow and enzymatic reactions in packed-bed reactors. They showed that the calculated properties agree well with the experimental data, thus confirming the general utility of pore network models for calculating the flow and other properties of a pore space, even in the case of a packing of particles, which is an unconsolidated porous medium. Earlier, Chu and Ng (1989) utilized a somewhat similar method.

A variation of the pore network model is one in which one utilizes the 3D image of the porous medium obtained by X-ray computed tomography. In that case, no simplifications in the microstructure need to be made, as the image can be used directly in the computations. One may use a lattice-Boltzmann method for simulating fluid flow or transport in the image, a method that was described in Chapter 9, or a numerical method such as the Galerkin finite-element method.

10.2

Exact Formulation and Perturbation Expansion

Consider, for example, diffusion (conduction) in heterogeneous porous media. In such porous media, the local (pore) diffusivity (conductivity) varies spatially, and the relevant equation to solve is

$$\frac{\partial C}{\partial t} = \nabla \cdot (D \nabla C), \quad (10.6)$$

where D denotes a spatially-varying diffusion coefficient (conductivity), and C denotes the concentration (temperature). Discretizing Eq. (10.6) by a finite-element or finite-difference method yields a *master equation*:

$$\frac{\partial C_i}{\partial t} = \sum_j W_{ij} [C_j(t) - C_i(t)], \quad (10.7)$$

where W_{ij} is related to the coefficients D_i and D_j at (grid) points i and j . For example, for a 1D system, $W_{ij} = (D_i + D_j)/(2\delta^2)$, where δ is the distance between i and j . Equation (10.7) can also be given a probabilistic interpretation: $C_i(t)$, when properly normalized, may be thought of as the probability that a diffusing species

would be at site i at time t if it commences its motion at the origin. According to the probabilistic interpretation, W_{ij} is the *transition rate* between sites i and j , that is, the probability of diffusing from i to j , taken to be nonzero only when sites i and j are nearest neighbors, if i and j represent the two end nodes of a pore in the pore network.

The exact and *explicit* solution of Eq. (10.7) is not available for any 2D or 3D model of disordered porous media or materials. Even if it were, it would contain more information than what is really needed or necessary. Therefore, one must resort to accurate approximations. Aside from numerical simulations, a powerful method of analyzing Eq. (10.7) is by perturbation expansion. In this section, we describe a general approach for developing perturbation expansion for such problems, which was first developed by Sahimi *et al.* (1983b).

10.2.1

Green Function Formulation and Perturbation Expansion

Taking the Laplace transform of Eq. (10.7) yields

$$\lambda \tilde{C}_i(\lambda) - \delta_{i0} = \sum_j W_{ij} [\tilde{C}_j(\lambda) - \tilde{C}_i(\lambda)] , \quad (10.8)$$

where λ is the Laplace transform variable conjugate to t , and the initial condition $C_i(0) = \delta_{i0}$ (i.e., a unit pulse injection at 0) has been used. A *uniform* network (or lattice) is now introduced in which all the transition rates are equal to $\tilde{W}_e(\lambda)$, with the site occupation probabilities being $\tilde{C}_i^0(\lambda)$. Therefore,

$$\lambda \tilde{C}_i^0(\lambda) - \delta_{i0} = \sum_j \tilde{W}_e(\lambda) [\tilde{C}_j^0(\lambda) - \tilde{C}_i^0(\lambda)] , \quad (10.9)$$

where the effective transition rate $\tilde{W}_e$ is yet to be determined. If one subtracts Eq. (10.8) from Eq. (10.9) (to eliminate the initial condition), one obtains, after some rearrangements,

$$\begin{aligned} (Z_i + \epsilon) [\tilde{C}_i(\lambda) - \tilde{C}_i^0(\lambda)] - \sum_j [\tilde{C}_j(\lambda) - \tilde{C}_j^0(\lambda)] \\ = - \sum_j \Delta_{ij} [\tilde{C}_i(\lambda) - \tilde{C}_j(\lambda)] , \end{aligned} \quad (10.10)$$

where Z_i is the coordination number of site i , $\epsilon = \lambda / \tilde{W}_e$, and $\Delta_{ij} = (W_{ij} - \tilde{W}_e) / \tilde{W}_e$. A *site* Green function $\mathcal{G}_{ik}$ is now introduced by the equation

$$(Z_i + \epsilon) \mathcal{G}_{ik} - \sum_j \mathcal{G}_{jk} = -\delta_{ik} . \quad (10.11)$$

Physically, $\mathcal{G}_{ik}$ is the response of the system at k if a unit “current” is injected into the network at i . In terms of $\mathcal{G}_{ik}$, Eq. (10.10) is rewritten as

$$\tilde{C}_i(\lambda) = \tilde{C}_i^0(\lambda) + \sum_j \sum_k \mathcal{G}_{ij} \Delta_{jk} [\tilde{C}_j(\lambda) - \tilde{C}_k(\lambda)] . \quad (10.12)$$

Equation (10.12) is an exact but implicit solution of Eq. (10.7) that is applicable to *any* network, irrespective of its dimensionality or topological structure. However, for topologically-disordered networks, for example, the Voronoi network (see Chapter 3), further progress requires a statistical treatment of the Green function appropriately coupled to the topological disorder of the network. The statistical treatment of $\mathcal{G}_{ik}$ was described by Sahimi and Tsotsis (1997). The derivation of the Green functions for regular lattices or networks was described by Sahimi *et al.* (1983b) and Sahimi (2003a). In particular, for a simple-cubic network in d dimensions ($Z = 2d$), the construction of the Green function is particularly straightforward. If we label integers $m_1, m_2, \dots, m_d$ by d , the relative positions of two sites i and j on the network separated by m_d bond lengths along the principal axis d , then the Green function $\mathcal{G}_{ij}$ is given by

$$\mathcal{G}_{ij} = \mathcal{G}(m_1, m_2, \dots, m_d) = -\frac{1}{2} \int_0^\infty \exp\left[-\frac{x(Z + \epsilon)}{2}\right] \prod_{i=1}^d I_{m_i}(x) dx. \quad (10.13)$$

Here, $I_{m_i}(x)$ denotes the modified Bessel function of order m_i . The Green functions for many other networks are given by Sahimi *et al.* (1983b) and Sahimi (2003a).

Equation (10.12) may also be rewritten in terms of the “flux”, $\tilde{Q}_{ij}(\lambda) = \tilde{C}_i(\lambda) - \tilde{C}_j(\lambda)$. We denote bonds with Greek letters, assign directions to them, and let $\gamma_{\alpha\beta} = (\mathcal{G}_{il} + \mathcal{G}_{jk}) - (\mathcal{G}_{jl} + \mathcal{G}_{ik})$ be a *bond* Green function, where i and l (j and k) are the network sites with tails (heads) of arrows on bonds α and β , respectively. Similar to $\mathcal{G}_{ij}$, the bond Green function $\gamma_{\alpha\beta}$ is the response of bond β of the network if a unit “current” is injected into bond α . In terms of $\gamma_{\alpha\beta}$, Eq. (10.12) is rewritten as

$$\tilde{Q}_\alpha(\lambda) = \tilde{Q}_\alpha^0(\lambda) + \sum_\beta \Delta_\beta \gamma_{\alpha\beta} \tilde{Q}_\beta(\lambda). \quad (10.14)$$

Similar to Eq. (10.12), Eq. (10.14) is also exact and expresses the flux $\tilde{Q}_\alpha$ in bond α in the disordered network as the sum of the flux $\tilde{Q}_\alpha^0$ in the same bond though in the uniform network, and the fluctuations in the flux through the bond that arise as the result of the heterogeneity of the network.

10.2.2

Self-Consistent Approximation

The above formulation is not complete until we devise a method for determining the effective transition rate $\tilde{W}_e$. This quantity is determined by the following *self-consistency* condition. Solve Eq. (10.14) for an arbitrary set of individual transition rates, that is, for arbitrary Δ_β . Select any bond α and require that

$$\langle \tilde{Q}_\alpha \rangle = \tilde{Q}_\alpha^0, \quad (10.15)$$

where $\langle \cdot \rangle$ denotes an averaging over all possible transition rates of all bonds. Equation (10.15) requires that the fluctuation in the flux in the bond (pore) α from its value in the effective (uniform) network vanish on average when the averaging is taken with respect to the disorder. If we substitute the effective transition rate obtained in Eq. (10.9) and invert the Laplace transform, we obtain

$$\frac{\partial C_i}{\partial t} = \sum_j \int_0^t W_e(t - \tau) [C_j(\tau) - C_i(\tau)] d\tau. \quad (10.16)$$

Thus, if we can determine $\tilde{W}_e$ exactly or approximately, we can replace the master equation (10.7) by a *generalized master equation*, Eq. (10.16). If, instead of a network, a continuum model is utilized and an analysis similar to the above is carried out, then the analog of Eq. (10.16) for a continuum model would be given by

$$\frac{\partial C}{\partial t} = \int_0^\infty D_e(t - \tau) \nabla^2 C(\mathbf{x}, \tau) d\tau, \quad (10.17)$$

where D_e is the analog of W_e , and $\mathbf{x}$ is a point in space. Equations (10.16) and (10.17) indicate that matching a heterogeneous porous medium (or any heterogeneous material for that matter) to a uniform medium induces *memory*. However, this does not necessarily imply that the long-time ($t \rightarrow \infty$) diffusivity of a species in the pore network depends on the time t . As shown below, whether the long-time diffusivity of a species in a porous medium is constant or depends on t is dependent upon the structure of the distribution of the heterogeneities of the medium, and particularly the spatial distribution of the local diffusivity D . Since the local diffusivities are controlled by the pores' size, this implies that the long-time behavior of D_e (or W_e) depends on the structure of the pore-size distribution.

10.2.3

Random Walks and Self-Consistent Approximation

As described earlier, Eq. (10.7) may be given a probabilistic interpretation in terms of the random walk or motion of a particle in the network. Such an interpretation provides a way of solving the diffusion or conduction problem. We will provide the complete details of the method later in this chapter, but we consider it briefly here, as it is related closely to what we have described so far.

One places a particle or diffusant at a site i of the network at position $\mathbf{r}_0$ at time $t = 0$. The particle is then allowed to move throughout the network. At every time step, the particle selects one of the bonds (pores) $i - j$ that are connected to site i at which it is currently residing with a probability proportional to the conductance of the bond. As the discretization of Eq. (10.6) demonstrated, the conductance is proportional to W_{ij} . The particle makes a jump (transition) to site j . Once at the new site, the particle selects its next step by the same method, makes another transition, and so on. This constitutes a random walk process.

Each time the particle makes a transition from one site to another, its current position $\mathbf{r}$ and $R^2(t) = (\mathbf{r} - \mathbf{r}_0)^2$ are calculated. One repeats this procedure for a large number of particles and a large number of time steps, and computes $\langle R^2(t) \rangle$, the mean-square displacement of the particles at time t , where the averaging is taken with respect to all the particles. It can be shown (see, for example, Hughes, 1995) that for a d -dimensional network (or continuum),

$$\langle R^2(t) \rangle = 2d D_e t. \quad (10.18)$$

Thus, if we compute $\langle R^2(t) \rangle$, the effective diffusivity D_e and, thus, the effective conductivity g_e of the system are determined. The Laplace transform of the mean-square displacement $\langle \tilde{R}^2(\lambda) \rangle$ of a diffusing particle is given by (Sahimi *et al.*, 1983b)

$$\langle \tilde{R}^2(\lambda) \rangle = \frac{Z}{\lambda^2} \tilde{W}_e(\lambda). \quad (10.19)$$

Thus, if W_e is accurately computed by the self-consistent approximation, Eq. (10.15), the mean-square displacements and, hence, the effective diffusivity via Eq. (10.18) will also be. The Einstein equation (see Chapter 3) that relates the effective conductivity and diffusivity is then invoked to obtain an estimate for the former.

10.2.4

Relation with Continuous-Time Random Walks

Kenkre *et al.* (1973) showed that a generalized master equation of the following form,

$$\frac{\partial P(\mathbf{l}, t)}{\partial t} = \int_0^t d\tau \varphi(t - \tau) \sum_{\mathbf{l}'} [p(\mathbf{l} - \mathbf{l}') P(\mathbf{l}', \tau) - p(\mathbf{l}' - \mathbf{l}) P(\mathbf{l}, \tau)], \quad (10.20)$$

can be put into a one-to-one correspondence with a continuous-time random walk (CTRW) on the same lattice. A CTRW is a random walk in which the walkers make an instantaneous step from one site to another, waits for a random time t , and then makes another instantaneous jump. The transition probability for a displacement from a site at $\mathbf{l}$ to site at $\mathbf{l}'$ is $p(\mathbf{l} - \mathbf{l}')$, and $\psi(t)$ represents the probability density function for the waiting times *between* the steps. The relation between $\psi(t)$ and $\varphi(t)$ in the Laplace transform space is given by $\tilde{\psi}(\lambda) = \tilde{\varphi}(\lambda)[\lambda + \tilde{\varphi}(\lambda)]^{-1}$. If we compare Eqs. (10.20) and (10.16), we see that $\tilde{\varphi}(\lambda) = Z \tilde{W}_e(\lambda)$. Thus, the waiting time density $\psi(t)$ is related to the effective transition rate $W_e(t)$ by (Sahimi *et al.* (1983b)

$$\tilde{\psi}(\lambda) = \frac{Z \tilde{W}_e(\lambda)}{\lambda + Z \tilde{W}_e(\lambda)}. \quad (10.21)$$

In view of Eq. (10.19), therefore, the waiting time density $\psi(t)$ is directly related to the mean-square displacement $\langle R^2(t) \rangle$. Such a relation between a random walk and a CTRW will be exploited in Chapter 11 in order to model dispersion in flow through porous media.

10.2.5

Effective-Medium Approximation

In practice, we cannot solve Eq. (10.15) for an *arbitrarily large* cluster of bonds, as the computations become too complex and, thus, we must develop approximate, but hopefully accurate, schemes, just as, as described in Chapter 9, it is not possible to solve a flow or transport problem in which many inclusions (such as spheres or ellipsoids) are embedded in an otherwise uniform or effective medium. The simplest of such approximations is the effective-medium approximation (EMA). Although the development of the EMA for the time-dependent transport phenomena described by Eqs. (10.6) and (10.7) began in the early 1980s, its counterpart for steady-state processes has a much longer history and goes back to the nineteenth century, when J.C. Maxwell developed his approximations that we described in Chapter 9. In fact, the EMA has its roots in the idea that in order to estimate the effective transport properties of a disordered medium, one may replace the disordered medium with a hypothetical homogeneous one that somehow mimics the behavior of the disordered medium. This is, of course, the approach that we used in order to arrive at Eqs. (10.12) and (10.14). If such a substitution is possible, the problem of calculating the effective transport properties of the homogeneous medium would be much simpler than the original problem, as the replacement greatly simplifies the problem.

There are two approaches for implementing this idea. Maxwell (1873) and Maxwell-Garnett (1904) developed an approach that is applicable to the case when isolated inclusions are embedded in a continuous matrix consisting of a single phase, that is, the dilute limit described in Chapter 9. The effective properties of the disordered medium are then derived by placing a sphere (or an ellipse) of the homogeneous or effective medium in the matrix. This is usually called the *average t-matrix* or the *nonself-consistent approximation*, and was briefly described in Chapter 9.

In the second approach developed by Bruggeman (1935)¹⁾, each inhomogeneity is embedded in the effective medium itself, the unknown properties of which are determined in such a way that the volume average over all the inhomogeneities yields no extra fields in the medium, that is, the average of the fluctuations vanishes. This is what is usually called the EMA, which was also briefly mentioned in Chapter 9 for the continuum models. In effect, the EMA is an ingenious way of transforming a many-body problem into a one-body problem. Bruggeman's EMA, which was derived for the continuum model of disordered media and mentioned in

1) *Dirk Anton George Bruggeman* was a Dutch physicist who received his Ph.D. at Utrecht. The modern theory of disordered media started with his 1935 paper, followed by five other papers published in 1935–1937.

Chapter 9, was rederived independently by Landauer (1952)²; see Landauer (1978) for a history of all the variations of the EMA.

In a practicable self-consistent approximation for estimating the effective flow and transport properties of the network models, one assigns the effective transition rate $\tilde{W}_e(\lambda)$ to all but a small cluster of bonds in the network, that is, $\Delta_\beta \neq 0$ only for a *finite* set of bonds, and proceeds as described by averaging over the transition rates of the bonds in the cluster in order to determine $\tilde{W}_e(\lambda)$. Clearly, one may also consider a cluster with two bonds, three bonds, and so on, each of which corresponds to a distinct level of approximation, and together they constitute a hierarchy of approximations. To obtain rapid convergence of the calculated effective properties to their true values in the original disordered network, it is important to choose a suitably symmetrical cluster of bonds with transition rates that are to be allowed to fluctuate, that is, be spatially distributed according to a statistical distribution. The simplest approximation, when only a single bond α has a transition rate W differing from $\tilde{W}_e(\lambda)$, represents the EMA. Figure 10.1 shows the schematic representation of the EMA concept. In this case, Eq. (10.15) simplifies to

$$\left\langle \frac{1}{1 - \gamma_{\alpha\alpha} \Delta_\alpha} \right\rangle = 1, \quad (10.22)$$

where $\gamma_{\alpha\alpha} = -2/Z + (2\epsilon/Z)\mathcal{G}(\epsilon)$, with $\mathcal{G}(\epsilon) = -\mathcal{G}_{ii}(\epsilon)$. Hence, if $f(W)$ is the probability density function of the transition rates, Eq. (10.22) yields

$$\int_0^\infty \frac{f(W)}{1 - \gamma_{\alpha\alpha} \Delta_\alpha} dW = 1. \quad (10.23)$$

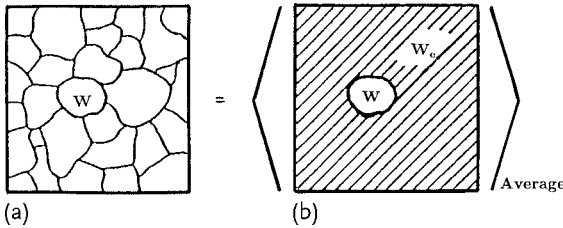


Figure 10.1 Schematic implementation of the EMA, transforming a disordered medium (a) to a homogeneous or uniform one (b).

2) *Rolf William Landauer* (1927–1999) was a German-American physicist who obtained his Ph.D. from Harvard University in 1950, and spent his entire career at NASA and IBM research laboratories. In addition to his contribution to the theory of inhomogeneous

media, he also made seminal contributions to the concepts of loss of information in irreversible electrical circuits that generate entropy and dissipated heat, which is essential to quantum information and quantum computing.

10.2.6

Effective-Medium Approximation and Percolation Disorder

A stringent test of any theory of transport in disordered media is its ability for accurately predicting the properties of a medium with percolation disorder. Thus, we consider a network in which a randomly-selected fraction p of the bonds are open – they allow diffusion and flow to occur and have finite conductivity (diffusivity) or permeability – while the rest are closed to diffusion (conduction) or flow. Thus, we use

$$f(W) = (1 - p)\delta_+(W) + p h(W), \quad (10.24)$$

where $h(W)$, the distribution of the transition rates of the open bonds, can be any normalized probability density function. In this limit, Eq. (10.21) becomes (Odagaki and Lax, 1981; Webman, 1981; Sahimi *et al.*, 1983b)

$$p \int_0^\infty \frac{h(W)}{1 - \frac{2}{Z} + \frac{2\mathcal{G}\epsilon}{Z} + 2(1 - \epsilon\mathcal{G})(W/\tilde{W}_e)/Z} dW = \frac{p - \frac{2}{Z} + \frac{2\mathcal{G}\epsilon}{Z}}{1 - \frac{2}{Z} + \frac{2\mathcal{G}\epsilon}{Z}}. \quad (10.25)$$

Equation (10.25) predicts that as long as $p > p_c$, $W_e = \tilde{W}_e(\lambda \rightarrow 0)$ is finite, so that Eq. (10.19) predicts that

$$\langle R^2(t) \rangle = Z W_e t, \quad (10.26)$$

which, together with Eq. (10.18), implies that $D_e = Z W_e / 2d$. Thus, diffusion in the network is described by the usual diffusion equation with an effective diffusion coefficient D_e .

What happens at $p = p_c$? As described in Chapter 3, at p_c , the sample-spanning cluster of the open or conducting bonds of the network is a fractal object, and the correlation length ξ_p of the network is divergent. Thus, $R = \sqrt{\langle R^2(t) \rangle}$ is always less than ξ_p and, because ξ_p is the only relevant macroscopic length scale of the network, diffusion is *not* Fickian. Indeed, it is not difficult to show that in this limit, Eq. (10.25) predicts that, for example, a simple-cubic network Sahimi *et al.* (1983b)

$$\tilde{W}_e(\lambda) \sim \left[\frac{p_c \mathcal{G}(0)}{(1 - p_c) h_{-1}} \right]^{\frac{1}{2}} \lambda^{\frac{1}{2}}, \quad (10.27)$$

where $h_{-1} = \int_0^\infty dW [h(W)/W]$. Therefore, Eq. (10.19) yields

$$\langle R^2(t) \rangle \sim \frac{12}{\pi^{\frac{1}{2}}} \left[\frac{p_c \mathcal{G}(0)}{(1 - p_c) h_{-1}} \right] t^{\frac{1}{2}}, \quad (10.28)$$

which indicates that the mean-square displacement of the diffusers grows with the time t *slower* than linearly. Therefore, the EMA correctly predicts that diffusion in a pore network at $p = p_c$ is *not* Fickian.

The non-Fickian nature of diffusion at the percolation threshold means that if we attempt to compute an effective diffusivity by writing $D_e = \langle R^2(t) \rangle / (6t)$, Eq. (10.28) would yield $D_e \propto 1/\sqrt{t}$. Therefore, $D_e \rightarrow 0$ as $t \rightarrow \infty$. That is, diffusion (or conduction) in a network at its percolation threshold is so slow that the long-time diffusivity is *zero*. The slowness of the diffusion process is, of course, due to the highly tortuous structure of the network and its poor connectivity at p_c . Note that even if the network is above its percolation threshold ($p > p_c$) though $R < \xi_p$, diffusion is still non-Fickian.

The difference between diffusion in a network below, at, and above its percolation threshold also explains why the $\tilde{W}_e$ that we used in Eq. (10.9) must depend on λ . Had we used a λ -independent W_e in Eqs. (10.9), (10.27) and (10.28) would have predicted that diffusion is *always* Fickian, *regardless* of whether the network is below, at, or above its percolation threshold. However, percolation theory of Chapter 3 taught us that for $p \leq p_c$, the effective diffusivity, conductivity, or permeability must be zero because no sample-spanning cluster of open bonds exists for macroscopic flow or transport. Non-Fickian diffusion is usually referred to as *anomalous diffusion* (Gefen *et al.*, 1983) or *fractal diffusion* (Sahimi *et al.*, 1983b). We will come back to anomalous diffusion shortly.

10.2.7

Steady-State Transport and Percolation Threshold

At steady state, $\epsilon = 0$, and $\gamma_{aa} = -2/Z$. After some rearrangements, Eq. (10.23) becomes

$$\int_0^\infty \frac{W - W_e}{W + \left(\frac{Z}{2} - 1\right) W_e} f(W) dW = 0. \quad (10.29)$$

For a d -dimensional simple-cubic network, $Z = 2d$ and Eq. (10.29) becomes

$$\int_0^\infty \frac{W - W_e}{W + (d - 1) W_e} f(W) dW = 0. \quad (10.30)$$

Equation (10.31) is *exactly* equivalent to Eq. (9.82).

At steady state, W is simply the bond conductance and, therefore, $f(W)$ represents the distribution of the bonds' (pores') conductance. Hence, $g_e = W_e$ is the effective conductivity of the network. Equation (10.29) was first derived by Kirkpatrick (1971, 1973). As described above and in Chapter 9, Eq. (10.30) had been derived much earlier by Bruggeman (1935) and Landauer (1952) for an isotropic continuum. Moreover, as described there, in the original Bruggeman–Landauer theory, in order to derive Eq. (10.30), a sphere was embedded in the effective medium similar to embedding a single bond in the effective network.

Despite its simplicity and our inability for including detailed information about the microstructure of the network in the formulation, the EMA can still predict a

nontrivial p_c . Setting $p = p_c$ and $W_e = 0$ in Eq. (10.29) yields

$$p_c = \frac{2}{Z} \quad (10.31)$$

for the bond percolation threshold of the network. The prediction of Eq. (10.31) is accurate for 2D networks, but not for the 3D ones. Recall from Chapter 3 that, in general, for a d -dimensional network, $p_c \simeq d/[Z(d-1)]$, which reduces to Eq. (10.31) for $d = 2$. Moreover, if we use $h(w) = \delta(W - W_0)$ in Eq. (10.24) and substitute the result in Eq. (10.29), we obtain

$$\frac{W_e}{W_0} = \frac{p - \frac{2}{Z}}{1 - \frac{2}{Z}}, \quad (10.32)$$

and, thus, W_e varies linearly with $p - p_c = p - 2/Z$. This implies that the EMA predicts that the critical exponent μ_p , which characterizes the power-law dependence of the conductivity of the network on p near p_c , is unity in both 2D and 3D networks. This is an incorrect result since $\mu_p(d=2) \simeq 1.3$ and $\mu_p(d=3) \simeq 2.0$. Even if we replace $\delta(W - W_0)$ with a more general distribution $h(W)$ as in Eq. (10.24), the exponent μ_p will still be one because, except in some special cases (see Chapter 3), the exponents are independent of $h(W)$ or any other detail of the network's structure.

10.2.8

Extensions of the Effective-Medium Approximation

One may also develop an EMA for the case in which there is a first-order chemical reaction (Sahimi, 1988b) in the pore network. Most of the above formulation remains intact. The main difference is that the Laplace transform variable λ is replaced by $\lambda + k_r$, where k_r is the reaction rate. In addition, most porous media are not completely random, but usually contain at least some short-range correlations. Hori and Yonezawa (1977) and Hilfer (1991b) modified the EMA to take into account the effect of such correlations. Sahimi (2003a) discussed many other extensions of the EMA.

10.2.9

Effective-Medium Approximation for Anisotropic Media

An important extension of the EMA is related to porous media and materials that are anisotropic. The extension was developed originally by Bernasconi (1974) for the square network, and later by Mukhopadhyay and Sahimi (2000) who developed a general framework for all networks. As described in Chapter 12, such an EMA may be used for estimating the effective permeability tensor of fracture networks that are usually anisotropic (Harris, 1990, 1992). Here, we briefly describe the extension and the results. More details are given by Mukhopadhyay and Sahimi (2000) and Sahimi (2003a).

We refer to the EMA for anisotropic networks as the anisotropic EMA (AEMA). The only difference with the isotropic case is that since we must compute d effective conductivities $g_{e\ell}$, one for each principal direction of the network, we need d self-consistency equations. Thus, Eq. (10.15) is written for each principal direction,

$$\langle Q_\alpha \rangle_\ell = (Q_\alpha^0)_\ell \quad \ell = 1, \dots, d. \quad (10.33)$$

It is then straightforward to show that the working equation for a single-bond AEMA is given by

$$\left\langle \frac{(\gamma_{\alpha\alpha})_\ell (g_\alpha - g_{e\ell})}{(\gamma_{\alpha\alpha})_\ell g_\alpha - [1 + (\gamma_{\alpha\alpha})_\ell] g_{e\ell}} \right\rangle = 0, \quad \ell = 1, \dots, d. \quad (10.34)$$

If the porous medium is characterized by direction-dependent conductance distributions $f_\ell(g_\alpha)$, then Eq. (10.34) becomes

$$\int_0^\infty \frac{g_\alpha - g_{e\ell}}{g_\alpha + S_\ell(g_{e1}, \dots, g_{ed})} f_\ell(g_\alpha) dg_\alpha = 0, \quad \ell = 1, \dots, d, \quad (10.35)$$

where

$$S_\ell = -\frac{1 + (\gamma_{\alpha\alpha})_\ell}{(\gamma_{\alpha\alpha})_\ell} g_{e\ell}. \quad (10.36)$$

Since $\gamma_{\alpha\alpha} = -2/Z$, we see that for isotropic networks, $S = (Z/2 - 1)g_e$. Thus, the main difference between the EMA and AEMA is in the quantity S_ℓ .

The bond Green function $(\gamma_{\alpha\alpha})_1$ in a principal direction of a square network can be shown to be given by

$$(\gamma_{\alpha\alpha})_1 = -\frac{1}{\pi} \left(\pi - 2 \arcsin \sqrt{\frac{g_{e2}}{g_{e1} + g_{e2}}} \right) = -\frac{1}{\pi} \left(\pi - 2 \arctan \sqrt{\frac{g_{e2}}{g_{e1}}} \right), \quad (10.37)$$

which reduces to $\gamma_{\alpha\alpha} = -1/2$ for the isotropic case ($g_{e1} = g_{e2}$), in agreement with $\gamma_{\alpha\alpha} = -2/Z = -1/2$ for $Z = 4$, which was computed earlier. Similarly,

$$(\gamma_{\alpha\alpha})_2 = -\frac{1}{\pi} \left(\pi - 2 \arctan \sqrt{\frac{g_{e1}}{g_{e2}}} \right), \quad (10.38)$$

which is obtained from Eq. (10.37) by cyclic permutation of the indices one and two. Then, for the square network in the principal direction $\ell = 1$, we have

$$S_1 = g_{e1} \frac{\arctan \left(\frac{g_{e2}}{g_{e1}} \right)^{\frac{1}{2}}}{\arctan \left(\frac{g_{e1}}{g_{e2}} \right)^{\frac{1}{2}}}, \quad (10.39)$$

and a cyclic permutation of the indices one and two leads to a formula for S_2 . Equation (10.39) was first given by Bernasconi (1974). In the case of a simple-cubic

network, however, the equations are more complex. Thus, we only give the final result,

$$S_1 = g_{e_1} \frac{\arctan [g_{e_1}^{-1} (g_{e_1} g_{e_2} + g_{e_1} g_{e_3} + g_{e_2} g_{e_3})]^{-\frac{1}{2}}}{\arctan [g_{e_1} (g_{e_1} g_{e_2} + g_{e_1} g_{e_3} + g_{e_2} g_{e_3})]^{-\frac{1}{2}}}, \quad (10.40)$$

and cyclic permutations of the indices lead to the corresponding formulae for R_2 , R_3 , S_2 and S_3 .

Another AEMA was developed by Koelman and de Kuijper (1997) for anisotropic materials that contain many components. However, the anisotropy that was considered in their work was due to shapes and orientations of the constitutive materials, for example, ellipsoidal grains in a powder or a porous material, which is different from what we describe here. The anisotropy of such porous materials persists even at the percolation threshold.

10.2.10

Continuum Models and Effective-Medium Approximation for Site-Disordered Networks

The continuum models of the type that were considered in Chapter 9 usually correspond to site-disordered networks, that is, those in which a site (and the bonds connected to it) is present with probability p . Recall from Chapter 3 that in site percolation, if a site of the network is removed, all the bonds that are connected to it are also deleted. In a continuum model, such as a packing of particles, the role of the sites is played by the particles. For this reason, one may develop an EMA for site-disordered networks. This task was undertaken by several groups, each developing an EMA for a particular type of network (Watson and Leath, 1974; Butcher, 1975; Bernasconi and Wiesmann, 1976; Joy and Strieder, 1978, 1979; Sahimi *et al.*, 1984). Sahimi (2003a) presented a general approach applicable to *all* site-disordered networks.

10.2.11

Accuracy of the Effective-Medium Approximation

In general, as Koplik (1981) showed, the EMA is very accurate if a network is not close to its percolation threshold, regardless of the structure of the distribution $h(W)$ in Eq. (10.24). As the example of the networks with percolation disorder demonstrated, the EMA predictions are more accurate for 2D networks than for the 3D ones. However, if percolation disorder is absent in a network, the predictions of the EMA are also accurate for 3D networks. The performance of the EMA near the percolation threshold may be improved systematically (Blackman, 1976; Turban, 1978; Ahmed and Blackman, 1979; Sheng, 1980; Sahimi, 1984; Sahimi and Tsotsis, 1997). To do so, a cluster of several bonds with random conductances is used in Eq. (10.22) instead of a single bond used in the formulation of the EMA, and then solved for W_e . A similar procedure is also used for the steady-state limit.

The most accurate results are obtained with the clusters that preserve the symmetry of the network. Erdős and Haley (1976) showed how different averaging schemes affect the accuracy of the EMA, and suggested an averaging scheme that improved the EMA's performance. Later in this chapter, we will describe another approach for improving the accuracy of the EMA. Detailed discussions of the merits and weaknesses of the EMA are given by Sahimi (2003a).

10.2.12

Effective-Medium Approximation for the Effective Permeability

Equation (10.29) may be used for estimating the effective permeability and diffusivity of porous media. Consider an effective-medium network in which each bond or pore has a conductance $g = W_e$. We fix the pressures at two opposite faces of the network so as to produce an average pressure gradient $\langle \nabla P \rangle$. The total volume flow rate Q crossing any plane perpendicular to the direction of the average pressure gradient is the sum of the individual volume flow rates in the bonds intersecting the plane. Each pore's volume flow rate is the pressure difference across it times g_e/μ , where μ is the fluid's viscosity. If we approximate the local pressure difference as the projection of the average pressure gradient along the bond length l , we find

$$Q = \sum \frac{g_e}{\mu} \langle \nabla P \rangle \cdot l. \quad (10.41)$$

If we divide Q by the area S of the plane, we obtain an average velocity that, when compared with the Darcy's law, yields an estimate of the effective permeability

$$K_e = g_e \left\langle \frac{1}{S} \sum l \cdot \mathbf{n} \right\rangle, \quad (10.42)$$

where $\mathbf{n}$ is a unit vector along the pressure gradient. If, however, the medium is statistically homogeneous and isotropic, any unit vector may be used. It should be pointed out that in this kind of approach, the pressure drop in the pore bodies (sites) is neglected. It is instead assumed that the pressure drops are due to the narrow pore throats (bonds) of the network. Koplik (1982) analyzed the case in which the pressure drops in both pore bodies and throats are taken into account. The analysis in this case is tedious and not given here. So long as the pore bodies are large and compact and the pore throats are long and narrow, the approximation of neglecting the pressure drops in the pore bodies is valid.

To calculate the effective diffusivity D_e , we use the following formula

$$D_e = \frac{1}{\pi d} \frac{g_e l}{\langle R^2 \rangle} \phi, \quad (10.43)$$

where $g_e = W_e$ is the EMA prediction of the conductance of the network, d is its dimensionality, l is the length of a pore throat, ϕ is the porosity of the network, R is a pore throat radius, and $\langle R^2 \rangle$ is an average with respect to the pore size distribution. It has been assumed that all the pore throats (bonds) have the same length.

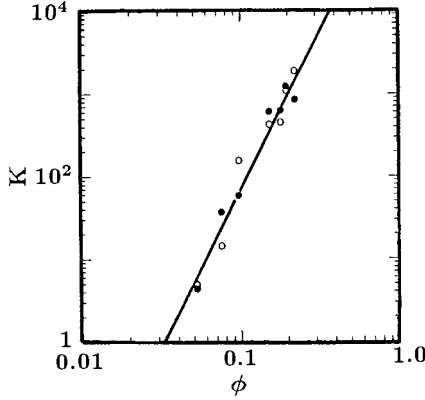


Figure 10.2 The logarithmic plot of dependence of the effective permeability K_e (in millidarcy) of Fontainebleau sandstone on the porosity. The slope of the straight line is 3.8 (after Doyen, 1988).

How accurate are the predictions of the EMA for the effective permeability and diffusivity of porous media? Koplik *et al.* (1984) analyzed a Massillon sandstone in detail, used a serial sectioning method to determine an equivalent random network to its pore space and, using this information, employed Eqs. (10.29) and (10.42) to calculate the effective electrical conductivity and permeability of the pore space. They found that the predicted K_e differs from the data by about one order of magnitude, while the predicted g_e differs by a factor of about 2. They attributed the difference to the fact that most sedimentary porous media, such as Massillon sandstone, are highly heterogeneous and anisotropic, properties that are not accurately taken into account by the EMA.

On the other hand, Doyen (1988) analyzed flow and transport properties of Fontainebleau sandstones and used Eqs. (10.29) and (10.42) to predict them. He found that both K_e and g_e could be predicted to be within a factor of 3. Figure 10.2 compares the predictions of the EMA with the data. The EMA has been utilized for predicting transport properties of many types of disordered materials, a list of which is too long to be given here (Sahimi, 2003a).

10.3

Anomalous Diffusion and Effective-Medium Approximation

As the discussions in the last section indicated, at the percolation threshold p_c , the mean-square displacement $\langle R^2(t) \rangle$ does not grow linearly with the time, rather it increases with t *slower* than linearly. To better characterize this phenomenon, we introduce a *random walk fractal dimension* D_w defined by

$$\langle R^2(t) \rangle \sim t^{\frac{2}{D_w}}, \quad (10.44)$$

so that Fickian diffusion corresponds to $D_w = 2$. It is then clear that the EMA, Eq. (10.28), predicts that so long as $h_{-1} < \infty$, one has a universal random walk fractal dimension, $D_w = 4$. However, if $h_{-1} = \infty$, then D_w will be nonuniversal. In particular, if $h(W) = (1 - \alpha)W^{-\alpha}$, where $0 < \alpha < 1$, then the EMA predicts that (Sahimi *et al.*, 1983b)

$$D_w = \frac{2 - \alpha}{2(1 - \alpha)} . \quad (10.45)$$

Note that a power-law conductance distribution $h(w)$ arises naturally in the Swiss-cheese model of porous media.

We already pointed out that even for $p > p_c$, diffusion may still be anomalous if $\langle R^2(t) \rangle^{1/2} \ll \xi_p$, where ξ_p is the correlation length of percolation (see Chapter 3). Therefore, one may define a time t_{co} such that for $t \ll t_{co}$, diffusion is anomalous, but Fickian for $t \gg t_{co}$. It can be shown that if h_{-1} is finite, then the EMA predicts that (Sahimi *et al.*, 1983b)

$$[t_{co}(p - p_c)^2]^{-1} \ln[t_{co}(p - p_c)] \sim 1 , \quad (10.46)$$

in 2D, and

$$t_{co} \sim (p - p_c)^{-2} \quad (10.47)$$

in 3D. Thus, in both cases, the crossover time *diverges* at $p = p_c$.

The divergence of t_{co} is caused by two factors. (1) As $p \rightarrow p_c$, the correlation length ξ_p diverges and, thus, the porous medium is inhomogeneous on a rapidly increasing length scale. (2) As $p \rightarrow p_c$, the number of dead-end bonds or sites also increases rapidly. Such bonds or sites slow down the diffusion process. The divergence of the time scale t_{co} for the crossover between Fickian and anomalous diffusion has an important implication for interpreting experimental data for the diffusivity in porous media in that one must ensure that the Fickian diffusion regime is reached before interpreting the experimental data as implying a constant effective diffusivity D_e .

10.3.1

Scaling Theory of Anomalous Diffusion

The accuracy of the predictions of the EMA for the time dependence of $\langle R^2(t) \rangle$ can be tested against the scaling theory of diffusion in disordered materials with percolation disorder. We consider diffusion on the sample-spanning percolation cluster. As described in Chapter 3, near p_c , one has

$$D_e \sim (p - p_c)^{\mu_p - \beta} \sim \xi_p^{-\theta} , \quad (10.48)$$

where $\theta = (\mu_p - \beta)/\nu$, and μ_p , β and ν are, respectively, the critical exponents of the conductivity, strength of the percolation, and the percolation correlation length

ξ_p . If $\langle R^2(t) \rangle^{1/2} \gg \xi_p$, then diffusion must be Fickian, so that the mean-square displacement grows linearly with the time t and, therefore,

$$\langle R^2(t) \rangle \propto D_e t \propto t(p - p_c)^{\mu_p - \beta} . \quad (10.49)$$

On the other hand, if the size of the cluster on which diffusion is taking place is finite, which happens when $p < p_c$, then after a long enough time, the diffusant has explored the cluster completely. Since the typical radius of such clusters is ξ_p , we must have

$$\langle R^2(t) \rangle \propto \xi_p^2 \propto |p - p_c|^{-2\nu} . \quad (10.50)$$

We may combine Eqs. (10.49) and (10.50) and write down a general scaling equation for the mean square displacement:

$$\langle R^2(t) \rangle^{\frac{1}{2}} = t^x f[(p - p_c)t^\gamma] . \quad (10.51)$$

For $p > p_c$ and long times, one must have $f(z) \sim z^{(\mu_p - \beta)/2}$ as $z \rightarrow \infty$ in order for Eq. (10.51) to be consistent with Eq. (10.49) and, thus, $x + \gamma(\mu_p - \beta)/2 = 1/2$. On the other hand, for $p < p_c$, the mean-square displacement must become independent of t , which would be the case if $f(z) \rightarrow z^{-x/\gamma}$ as $z \rightarrow -\infty$, implying that $x/\gamma = \nu$. Thus, solving for x and γ , we obtain $x = \nu/(2\nu + \mu_p - \beta)$ and $\gamma = x/\nu$. Since $\langle R^2(t) \rangle \sim t^{2x}$, we obtain

$$\langle R^2(t) \rangle \sim t^{\frac{2\nu}{2\nu + \mu_p - \beta}} , \quad (10.52)$$

which, when compared with Eq. (10.44), implies that the random walk fractal dimension is given by

$$D_w = 2 + \frac{\mu_p - \beta}{\nu} = 2 + \theta . \quad (10.53)$$

This result is usually attributed to Gefen *et al.* (1983). Equation (10.53) may also be derived by the following more intuitive method. Since fractal or anomalous diffusion occurs only if $\langle R^2(t) \rangle^{1/2} \ll \xi_p$, one can replace ξ_p with $\langle R^2(t) \rangle^{1/2}$ (since for $\langle R^2(t) \rangle^{1/2} \ll \xi_p$, the only relevant length scale of the network is $\langle R^2(t) \rangle^{1/2}$) and write $D_e \sim \langle R^2(t) \rangle^{-\theta/2}$. On the other hand, Eq. (10.18) implies that $D_e \sim d\langle R^2(t) \rangle/dt \sim \langle R^2(t) \rangle^{-\theta/2}$, which after integration, yields $\langle R^2(t) \rangle \sim t^{2/(2+\theta)}$ and, thus, we recover Eq. (10.53).

Since, as discussed in Chapter 3, the exponents μ_p , β and ν depend on the space dimensionality d , the random walk fractal dimension D_w also varies with d , whereas the EMA always predicts that $D_w = 4$. However, using the numerical values of the exponents, one finds that $D_w(d = 3) \simeq 3.8$, so that the actual value of D_w for 3D media is only about 5% smaller than the EMA prediction, $D_w = 4$. From the scaling equation (10.51), one also obtains the power law that governs the time t_{co} at which a crossover between fractal and Fickian diffusion takes place. It is clear that $t_{co} \sim \xi_p^{D_w}$ and, therefore

$$t_{co} \sim (p - p_c)^{-(2\nu + \mu_p - \beta)} . \quad (10.54)$$

Equation (10.54) predicts that in 3D, $t_{\text{co}} \sim (p - p_c)^{-3.34}$, which should be compared with the prediction of the EMA, Eq. (10.47). Thus, once again, the EMA prediction is *qualitatively* correct.

Thus far, the results pertain to diffusion on the sample-spanning percolation cluster. One can also consider diffusion on *all* the clusters (sample-spanning or not), which is relevant to diffusion in porous media. In this case, $\langle R^2(t) \rangle$ must be averaged over all the clusters. As described in Chapter 3, the probability that a site of a network that belongs to a cluster of size s is sn_s , where n_s is the number of clusters of size s . If R_s^2 is the mean-square displacement of a particle on a cluster of size s , then

$$\langle R^2(t) \rangle (t \rightarrow \infty, p < p_c) = \sum_s sn_s R_s^2 \propto |p - p_c|^{\beta - 2\nu}. \quad (10.55)$$

Because diffusion occurs on all the clusters, then, above p_c , the diffusivity is directly proportional to g_e , the effective conductivity, and, therefore, $D_e \sim (p - p_c)^{\mu_p}$ and $\langle R^2(t) \rangle \sim D_e t \sim t(p - p_c)^{\mu_p}$, which, when combined with Eq. (10.55), may be rewritten as a single scaling equation,

$$\langle R^2(t) \rangle^{\frac{1}{2}} \propto t^k f[(p - p_c)t^\gamma]. \quad (10.56)$$

Similar to Eq. (10.51), the exponents k and γ may be obtained by demanding that the scaling function $f(z)$ must be consistent with Eq. (10.56), $p > p_c$ ($z \rightarrow +\infty$) and $p < p_c$ ($z \rightarrow -\infty$). One then obtains $k = (\nu - \beta/2)/(2\nu + \mu_p - \beta)$ and $\gamma = (2\nu + \mu_p - \beta)^{-1}$. Since $\langle R^2(t) \rangle \sim t^{2k}$, we obtain

$$\langle R^2(t) \rangle \sim t^{\frac{(2\nu - \beta)}{(2\nu + \mu_p - \beta)}}, \quad (10.57)$$

which should be compared with Eq. (10.52). Equation (10.57) predicts that in 3D, one must have $\langle R^2(t) \rangle \sim t^{0.33}$ if we use the numerical values of the exponents in 3D, which should be compared with the prediction of Eq. (10.52), $\langle R^2(t) \rangle \sim t^{0.527}$. Clearly, diffusion on the finite clusters further slows down the overall transport process. Note that since the EMA replaces a heterogeneous network by a uniform one, it cannot distinguish between diffusion on the sample-spanning cluster and diffusion on all the clusters. Many other aspects of anomalous diffusion are discussed in detail by Havlin and Ben-Avraham (1987) and Haus and Kehr (1987).

10.3.2

Experimental Test of Anomalous Diffusion

Equations (10.52) and (10.57) and the predicted values of D_w have been tested and confirmed by numerical simulations of diffusion in percolation clusters. However, an interesting experimental verification of the above results was presented by Knackstedt *et al.* (1995) who prepared a porous material, a ternary microemulsion comprised of three components, didodecyl dimethyl ammonium bromide, water, and cyclohexane, which was a bicontinuous water-oil system. However, by tuning

the volume fraction of the three components, it could undergo, at high water contents, a structural transition to disconnected water droplets in oil, that is, the water phase would undergo a percolation transition, thus allowing measurement of the various mechanical and transport properties, including the diffusivity. Knackstedt *et al.* (1995) measured water self-diffusion D_e in such a porous medium by pulsed field gradient spin-echo technique at a controlled temperature of 25 ± 0.5 °C. By varying the length of the gradient pulse $\mathcal{P}$ and maintaining a constant gradient pulse interval Δ , the diffusion coefficient was measured. The decay I of the echo density is given by

$$I = I_0 \exp \left[-D_e G^2 \mathcal{P}^2 \gamma^2 \left(\mathcal{P} - \frac{\Delta}{3} \right) \right], \quad (10.58)$$

where G is the gradient strength, γ is the gyromagnetic ratio of the observed nucleus (^1H in this case), and I_0 is the signal intensity in the absence of a gradient pulse. The gradient strength was calibrated with a sample of H_2O for which D_e was known.

Diffusion in the porous material that Knackstedt *et al.* studied corresponds to the case in which the transport occurs on all the clusters and, therefore, the relevant equation is Eq. (10.57). Anomalous diffusion occurred for intermediate values of the water volume fractions, 13.5–14.1%. Since $D_e \sim d\langle R^2(t) \rangle / dt \sim t^{2k-1}$, a logarithmic plot of D_e versus t should be a straight line with a slope $2k - 1$. Knackstedt *et al.*'s data yielded $2k \simeq 0.3 \pm 0.1$, in good agreement with the prediction of the scaling theory, $2k \simeq 0.33$.

10.3.3

The Governing Equation for Anomalous Diffusion

Since anomalous diffusion results in a time-dependent effective diffusivity, the classical diffusion equation with a constant diffusivity no longer describes the diffusion process. Therefore, an important question has been the functional form of the equation that governs anomalous diffusion, a problem that has been studied for many years. For simplicity, we consider the problem in radial geometry and consider $P(r, t)$, the probability of finding the diffusant at position r at time t . The governing equation for anomalous diffusion is given by (Metzler *et al.*, 1994; Zeng and Li, 2000)

$$\frac{\partial^{\frac{2}{D_w}} P(r, t)}{\partial t^{\frac{2}{D_w}}} = \frac{1}{r^{D_s-1}} \frac{\partial}{\partial r} \left[r^{D_s-1} \frac{\partial P(r, t)}{\partial r} \right]. \quad (10.59)$$

The quantity $D_s = 2D_f/D_w$, where D_f is the fractal dimension of the medium (the fractal dimension of the sample-spanning percolation cluster), is called the *spectral* or *fracton* dimensionality. The derivative on the left-hand side of Eq. (10.59) is called *fractional derivative* (Hilfer, 2000) and is defined by

$$\frac{\partial^\gamma P(r, t)}{\partial t^\gamma} = \frac{1}{\Gamma(1-\gamma)} \frac{\partial}{\partial t} \int_0^t \frac{P(r, \tau)}{(t-\tau)^\gamma} d\tau, \quad (10.60)$$

where $\Gamma(x)$ is the gamma function. Using the Laplace transform, the asymptotic solution of Eq. (10.59) is obtained with the result being

$$P(r, t) \sim t^{-\frac{D_s}{2}} \exp \left[-c \left(\frac{r}{t^{\frac{1}{D_w}}} \right)^{\frac{D_w}{(D_w-1)}} \right], \quad (10.61)$$

where c is a constant. It can now be easily shown that

$$\langle R^2(t) \rangle = \int_0^t r^2 P(r, t) r^{D_t-1} dr \sim t^{\frac{2}{D_w}}, \quad (10.62)$$

in agreement with Eq. (10.44).

10.4

Archie's Law and the Effective-Medium Approximation

A useful empirical equation for sedimentary rocks is Archie's law (Archie, 1942),

$$g_e = g_f \phi^m, \quad (10.63)$$

where g_f is the electrical conductivity of a fluid (such as brine) saturating a porous medium, and g_e is the effective electrical conductivity of the medium. The exponent m has been found to vary anywhere between 1.3 and 4, depending upon consolidation and other factors. Archie's law has been found to hold even for igneous rocks (Brace *et al.*, 1968; Brace and Orange, 1968). However, Archie's law may take a more complex form for clayey or shaly porous media because, for example, the clays that are capable of ion exchange, can complicate the conduction mechanisms. Note that Archie's law implies that the fluid phase remains connected at all saturations, that is, the critical porosity or the percolation threshold is zero.

There have been many attempts to derive Archie's law in order to understand its origin and physical significance; here, we describe briefly the works. Sen (1981, 1984), Sen *et al.* (1981), Mendelson and Cohen (1982), and Yonezawa and Cohen (1983) showed that a modification of the EMA may be used to derive Archie's law. This version of the EMA was called the *self-similar* EMA (see Chapter 9) due to the assumption that a rock grain is coated with the conducting fluid that includes coated rock grains, with the coating at every level consisting of other coated grains.

Consider, first, the standard EMA, Eq. (10.29). With a binary distribution, $f(W) = p \delta(W - g_f) + (1 - p) \delta(W - g_r)$ and $g_e = W_e$, it becomes

$$g_e = \frac{-A \pm \sqrt{A^2 + 4(\gamma^{-1} - 1)g_f g_r}}{2(\gamma^{-1} - 1)}, \quad (10.64)$$

where $\gamma^{-1} = Z/2$ in the EMA for pore networks, Eq. (10.29), $\gamma^{-1} = d$ for a d -dimensional continuum, as in Eq. (10.30), and

$$A = g_f - (\gamma^{-1} - 1)g_r - \gamma^{-1}(g_f - g_r)p,$$

where g_r is the rock conductivity. Equation (10.64) has two solutions, but only one of them is physically meaningful. In the limits $p \rightarrow 0$ and $p \rightarrow 1$, we obtain

$$g_{(1)} = g_f \left[1 + (1 - p) \gamma^{-1} \frac{g_r - g_f}{g_r + (\gamma^{-1} - 1)g_f} \right], \quad (10.65)$$

$$g_{(2)} = g_r \left[1 + p \gamma^{-1} \frac{g_f - g_r}{g_r + (\gamma^{-1} - 1)g_r} \right]. \quad (10.66)$$

The basis for the self-similar EMA is as follows. One starts with the pure conducting fluid, replaces small portions of it by pieces of rock step by step, and applies the EMA to the mixture at each step. Assume that $g_e^{(i)}$ is the conductivity of the mixture (of rock and fluid) at step i , and replace a small volume Δq_i of the medium by grains of rock. Equations (10.29) and (10.30) yield

$$\frac{g_e^{(i)} - g_e^{(i+1)}}{g_e^{(i)} + (\gamma^{-1} - 1)g_e^{(i+1)}} (1 - \Delta q_i) + \frac{g_r - g_e^{(i+1)}}{g_r + (\gamma^{-1} - 1)g_e^{(i+1)}} \Delta q_i = 0. \quad (10.67)$$

If Δq_i is small enough, Eq. (10.67) yields

$$g_e^{(i+1)} = g_e^{(i)} \left[1 + \gamma^{-1} \frac{g_r - g_e^{(i)}}{g_r + (\gamma^{-1} - 1)g_e^{(i)}} \Delta q_i \right]. \quad (10.68)$$

Note that, by denoting the volume fraction of the fluid at the i th stage by p_i , we have $\Delta p_i q_i = p_i - p_{i+1}$. If we use Eq. (10.68) repeatedly, we obtain, in the limit, $g_r = 0$,

$$g_e^{(n+1)} = \sum_{i=1}^n \left[1 + \frac{\gamma^{-1}}{\gamma^{-1} - 1} \frac{p_{i+1} - p_i}{p_i} \right] g_e^{(0)}, \quad (10.69)$$

where $g_e^{(0)} = g_f$ and $p_0 = 1$ ($p_0 = 1$ corresponds to a pure fluid). In the limit, $\Delta q_i \rightarrow 0$, Eq. (10.67) becomes a differential equation:

$$\frac{dg_e^{(i)}}{g_e^{(i)}} = - \frac{dp_i}{p_i} \frac{g_f - g_e^{(i)}}{g_f + (\gamma^{-1} - 1)g_e^{(i)}} \gamma^{-1}, \quad (10.70)$$

which, after integrating and using the boundary condition that, $g_e^{(i)} = g_f$ for $p_i = 1$, yields Eq. (10.63) with

$$m = \frac{\gamma^{-1}}{\gamma^{-1} - 1}. \quad (10.71)$$

Thus, if we interpret p as the porosity of the porous medium, then the EMA produces Archie's law. Equation (10.71) also indicates that, consistent with considerable experimental data, the exponent m of Archie's law is not universal, but depends on the connectivity of the porous medium.

What is the geometrical interpretation of the derivation process? In the context of the pore network models, an interesting interpretation was presented by Yonezawa

and Cohen (1983) that may be summarized as follows. At the i th stage, every bond (pore) has a conductivity $g_e^{(i)}$ and, then, a small fraction Δq_i of the bonds is replaced by a resistor with conductivity g_r . Then, the EMA is used to estimate the effective conductivity of the new mixture of the rock and fluid. This is equivalent to inserting a resistor parallel to the original one in each bond. The conductivity $g_a^{(i)}$ of the added resistor should be

$$g_a^{(i)} = \gamma^{-1} g_e^{(i)} \frac{g_2 - g_e^{(i)}}{g_2} + (\gamma^{-1} - 1) g_e^{(i)} \Delta q_i, \quad (10.72)$$

if the original resistor belongs to the host medium; otherwise, it is given by Eq. (10.68). The implication is that even when $g_r = 0$, the application of the EMA at each stage makes the link between the nodes of the pore network conducting due to the addition of a parallel conducting resistor and, thus, there is always a sample-spanning cluster of conducting bonds. Translating this for the rock-fluid system, it implies that this procedure guarantees the continuity of the fluid phase and the granularity of the rock grains.

Note that in the original derivation of Sen *et al.* (1981), the Archie exponent m was found to be $3/2$, which corresponds to $d = 3$ in the EMA for a continuum, Eq. (10.30) (or $Z = 6$ in the EMA for pore networks, Eq. (10.29)), which corresponds to spherical grains. For nonspherical grains, $m > 3/2$, but under certain circumstances, one may even have $m < 3/2$. Mendelson and Cohen (1982) gave $m = 1/3 \sum_i (1 - A_i^*)^{-1}$, where A_i^* is the depolarization factor given by Eq. (9.81) with $\sum_i A_i^* = 1$. Bussian (1983) generalized the self-similar EMA to include finite rock conductivity g_r , and fitted the resulting formula to the data, assuming that m and g_r are adjustable parameters. He found $m \geq 3/2$ in almost all the cases that he considered, and argued that this was due to clay having a finite value g_r and, as clay particles are usually flat, they increase m (Mendelson and Cohen, 1982).

One major drawback of the derivation of Archie's law based on the EMA is that it pertains only to a microstructure in which the solid component is disjoint. This difficulty was addressed by Sheng (1990) who generalized the self-similar EMA to a *three-component* mixture (see also Chapter 9), with the third component being the *cement material*. Component one, the starting phase, is composed of a mixture of the fluid and cement material. Sheng (1990) showed that the self-similar EMA with three components reproduces Archie's law with

$$m = \frac{5 - 3A_g^*}{3 \left[1 - (A_g^*)^2 \right]}, \quad (10.73)$$

where A_g^* is the depolarization coefficient of the grains, but with the added feature that the solid grains also remain connected.

Although the self-similar EMA is successful in providing a derivation of Archie's law, its use for understanding various properties of rock is not without conceptual difficulty. Generally speaking, rock porosities are less than 0.4. This is far from the dilute limit in which the assumptions of the EMA and the model can be justified (recall that Eq. (10.68) is valid only in the dilute limit). If the porosity is low,

then the grains are in close contact with one another and the interaction between them is important. As described both in Chapter 9 and earlier in the present chapter, such interactions cannot be taken correctly into account by the EMA. In fact, Milton (1984) showed that the self-similar EMA accounts for the interactions correctly only in the special case in which grains of any given size are surrounded by much smaller grains, and grains of the same size are far separated from each other. This is hardly the case in natural porous media. Moreover, rocks with very similar grains may have very different values of the Archie exponent m , and rocks with very dissimilar grains can have very similar values of m . Such features cannot be explained with the self-similar EMA. Hilfer (1991a) presented an alternative derivation of Archie's law based on a percolation model. His result, $m = 1 + \mu_p$ (where μ_p is the conductivity exponent defined in Chapter 3), indicates that m is universal unless μ_p is nonuniversal, which is the case only in certain model porous media (see Chapter 3).

Wong *et al.* (1984) presented another derivation of the Archie's law, with the caveat that the Archie exponent m depended on the skewness of the pore size distribution. Their experiments with fused-glass beads and natural porous media indicated that m is larger if there are wider fluctuations of the pore sizes, which their model also predicted. Moreover, Wong *et al.* (1984) showed that their model predicts that

$$K_e \sim \phi^{m'}, \quad (10.74)$$

where $m' = 2m$. Equation (10.74) is consistent with the empirical Kozeny–Carman correlation (see Chapter 9). Note that if, for example, $m = 3$, we obtain $m' = 6$, consistent with the experimental observations (Wyllie and Rose, 1950; Timur, 1968) that if the effective permeability K_e is to be related to the porosity ϕ by a power law, then its exponent must be large. Indeed, the data of Rumpf and Gupte (1971) for random bead packs suggest $m' \simeq 5.5$.

10.5

Renormalization Group Methods

The discussions so far should have made it clear that the basis for developing the EMA is that fluctuations in the potential (pressure, voltage, temperature, and concentration) field are small because, as discussed in Section 10.1, in deriving the EMA, we require the average of the fluctuations in the field to be zero. If, however, the fluctuations are large, such as, for example, when a porous medium is near its critical porosity or percolation threshold, or when a porous medium has a broad distribution of the (hydraulic, diffusive, thermal, or electrical) conductances, the EMA breaks down. In such cases, a position-space renormalization group (PSRG) method is more appropriate because, in averaging the properties of the system, the PSRG method takes into account the properties of the *pre-averaged* network. It also predicts nonanalytic power laws – with exponents that are not unity or rational

numbers – for flow and transport properties near the percolation threshold, a distinct advantage over the EMA that always predicts the exponents of the power laws to be unity.

To describe the PSRG method, we again use the analogy between fluid flow and conduction. Consider, for example, a square or a cubic network in which each bond is conducting with probability p . The conductance can be hydraulic, diffusive, electrical, or thermal. The idea in the PSRG method is that since the actual network is so large that its properties cannot be computed exactly (except by resorting to numerical calculations), we partition it into $b \times b$ or $b \times b \times b$ blocks – called the RG cells – where b is the number of bonds in any direction, and calculate their effective properties that are hopefully representative of the properties of the original network. However, the effective properties of the RG cells are calculated in a particular way that we describe shortly. The shape of the RG cell can be selected arbitrarily, but it should be chosen in such a way that it preserves the symmetries of the original network as much as possible. For example, since the square network is self-dual, and because self-duality plays an important role in its percolation, flow, and transport properties, we use self-dual cells to represent the network (Bernasconi, 1978). Note that the dual of a network is obtained by connecting the center of each block to those of its nearest neighbors. Figure 10.3 shows examples of the RG cells with $b = 2$ in the square and simple-cubic networks, where the 2D cell is self-dual.

The next step in the PSRG method is to replace each RG cell with one bond in each principal direction (see Figure 10.3). If in the original network each bond conducts with probability p , then the bonds that replace the cells would be conducting with probability $p' = R(p)$, where $R(p)$ is called the *RG transformation* and represents the *sum* of all the probabilities of *all* the conducting configurations of the RG cell. To compute $R(p)$, we proceed as follows. Since we are interested in flow and transport in the original network, which is supposed to be represented by the RG

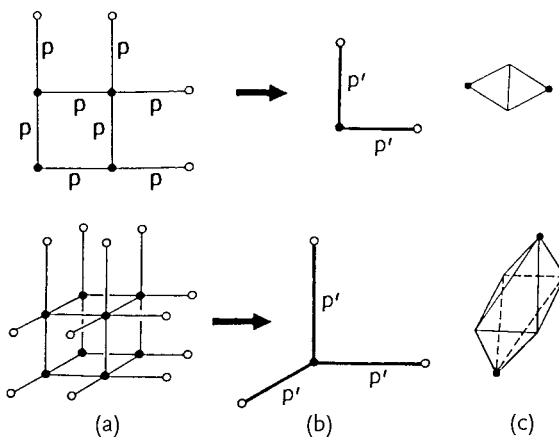


Figure 10.3 RG cells (a), their renormalized configurations (b), and the equivalent circuits (c). Here, $b = 2$.

cell, we solve for the flow and transport problems in each cell by applying a fixed potential gradient across the RG cell in a given direction. For example, the 2×2 RG cell of Figure 10.3 is equivalent to the circuit shown there, which is usually called the *Wheatstone bridge*. Thus, for the $b = 2$ cell, we only need to deal with a circuit of five bonds, and for the $2 \times 2 \times 2$ cell, we construct an equivalent 12-bonds circuit (see Figure 10.3). To compute $R(p)$, we determine all the conducting configurations of such circuits (i.e., all the configurations of the conducting bonds that form a sample-spanning cluster across the circuit), with some bonds conducting and some insulating. Thus, for the 2×2 RG cell, we obtain

$$p' = R(p) = p^5 + 5p^4q + 8p^3q^2 + 2p^2q^3, \quad (10.75)$$

where $q = 1 - p$. It is easy to see how Eq. (10.75) is derived: There is only one conducting RG cell configuration with all the five bonds conducting (probability p^5), five conducting configurations with four bonds conducting and one bond insulating (probability $5p^4q$), and so on.

As discussed in Chapter 3, the sample-spanning cluster at p_c is self-similar, implying that the RG transformation should remain invariant at p_c . The invariance of $R(p)$ should also be true at $p = 1$ and $p = 0$ because under any reasonable RG transformation, full (all bonds conducting) and empty (no bond conducting) networks should be transformed to full and empty networks again. The points $p = 0$, 1, and p_c are called the *fixed points* of the transformation, and are denoted by p^* . Since the RG transformation should not change anything at these points, the implication is that at the fixed points, the probability (p) of having a conducting bond in the RG cell and that in the renormalized cell ($p' = R(p)$) should be the same. Thus, the fixed points should be the solution of the polynomial equation

$$p^* = R(p^*), \quad (10.76)$$

and indeed this equation usually has three roots that are, $p^* = 0$, $p^* = 1$, and $p_c = p^*$, where p^* is the prediction of the RG transformation for the percolation threshold p_c . For the RG cells of Figure 10.3, we obtain $p^* = 1/2$ for both 2×2 and 3×3 cells, the exact result. In fact, it can be shown (Bernasconi, 1978) that the RG transformation for such 2D self-dual cells of *any* size always predicts $p_c = 1/2$. With the $2 \times 2 \times 2$ RG cell we obtain $p^* \simeq 0.208$, which should be compared with the numerical estimate, $p_c \simeq 0.249$.

So far, the PSRG method seems simple enough that the small-cell calculations can be done even on the “back of the envelope”, with the results being reasonably accurate. One may guess that one should calculate $p_c = p^*$ for several cell sizes b and then use the finite-size scaling (see Chapter 3) according to which

$$p_c - p_c(b) \sim b^{-\frac{1}{\nu}}, \quad (10.77)$$

where p_c is the true percolation threshold of the network, and ν is the critical exponent of percolation correlation length ξ_p , to extrapolate the results to $b \rightarrow \infty$ and estimate p_c . Indeed, Reynolds *et al.* (1980) used this method and obtained very accurate estimates of site and bond percolation thresholds of the square network.

However, as always, life is more complex than we would like it to be. Ziff (1992) showed that as $b \rightarrow \infty$, the probability $R(p_c)$ approaches a *universal* value of $1/2$ for *all* the 2D networks. This implies that, *asymptotically*, Eq. (10.76) is *wrong*, because p_c is *not* universal and depends on the structure of the network, whereas $R(p_c)$ is universal (as $b \rightarrow \infty$). Ziff's discovery did not violate the universality of $R(p_c)$; it only fixed its value. Stauffer *et al.* (1994) showed that $R(p_c)$ is also universal in 3D.

We may ask then, how can one use an RG transformation to estimate p_c ? Although, due to Ziff's discovery, the answer to this question during 1992–1994 was not clear, with most of the results that had been obtained (based on Eq. (10.76)) in the 1970s and early 1980s in doubt, Sahimi and Rassamdana (1995) showed that, although $R(p_c)$ is universal, for *any* $0 < \alpha < 1$ the equation

$$R(p_c) = \alpha, \quad (10.78)$$

always provides an estimate of p_c in $(0,1)$. Therefore, one can determine $R(p_c)$ for a series of RG cells of increasing linear sizes b , solve Eq. (10.78) for the corresponding values of p_c , and use Eq. (10.77) to extrapolate the results to $b \rightarrow \infty$. In other words, although Eq. (10.76) is *wrong theoretically*, it still yields the correct estimates of p_c . Moreover, it appears that among all the possible values, $\alpha = p_c$ still provides the fastest convergence to the true value of p_c . In addition, one has a bonus in that the critical exponent ν of the percolation correlation length is estimated based on $R(p_c)$. Suppose that the percolation correlation length in the original and renormalized network (the RG cells) are ξ_p and ξ'_p , respectively. Since each bond of the RG cells is replaced by another bond b times its length, we must have

$$\xi'_p = \frac{1}{b} \xi_p. \quad (10.79)$$

On the other hand, $\xi_p \sim (p - p_c)^{-\nu}$ and, due to the universality of the percolation exponents, $\xi'_p \sim [R(p) - R(p_c)]^{-\nu}$. If we linearize $R(p)$ and $R(p_c)$ around $p_c = p^*$, and use Eq. (10.79), we obtain

$$\nu = \frac{\ln b}{\ln \lambda}, \quad (10.80)$$

where $\lambda = dR(p)/dp$, evaluated at $p = p^*$. For example, the 2×2 and 3×3 self-dual RG cells yield $\nu \simeq 1.43$ and 1.38 , respectively, which should be compared with the exact value $\nu = 4/3$. For the $2 \times 2 \times 2$ cell of Figure 10.3, one obtains $\nu \simeq 1.03$, which should be compared with the numerical estimate, $\nu \simeq 0.88$ (see Chapter 3).

The PSRG approach may also be used for calculating the conductivity of a random conductance network. One begins with the original probability density function $f_0(g)$ for the bond conductances g of the RG cell and replaces it with a new distribution $f_1(g)$, the probability distribution for the conductance of a bond in the renormalized RG cell, which is calculated by determining the equivalent conductance of the RG cell. Thus, for an n -bond RG cell, one obtains a recursion relation relating $f_1(g)$ to $f_0(g)$:

$$f_1(g) = \int f_0(g_1) dg_1 f_0(g_2) dg_2 \dots f_0(g_n) dg_n \delta(g_p - g'), \quad (10.81)$$

where $g_1, \dots, g_n$ are the conductances of the n bonds of the RG cell, and g' is the equivalent conductance of the RG cell. For example, for the 5-bond cell of Figure 10.3, one has

$$g' = \frac{g_1(g_2g_3 + g_2g_4 + g_3g_4) + g_5(g_1 + g_2)(g_3 + g_4)}{(g_1 + g_4)(g_2 + g_3) + g_5(g_1 + g_2 + g_3 + g_4)}. \quad (10.82)$$

and if $f_0(g) = (1 - p)\delta(g) + p\delta(g - g_0)$, then (using Eq. (10.81))

$$\begin{aligned} f_1(g) = & [1 - R(p)]\delta(g) + 2p^3q^2\delta\left(g - \frac{1}{3}g_0\right) + 2p^2(1 + 2p)q^2\delta\left(g - \frac{1}{2}g_0\right) \\ & + 4p^3q\delta\left(g - \frac{3}{5}g_0\right) + p^4\delta(g - g_0), \end{aligned} \quad (10.83)$$

which is already more complex than $f_0(g)$. One now iterates Eq. (10.81) to obtain a new distribution $f_2(g)$ by substituting $f_1(g)$ into its right side. The iteration process should continue until a distribution $f_\infty(g)$ is reached, the shape of which does not change under further iterations. This is called the *fixed-point distribution* and the conductance of the original network is simply an average of $f_\infty(g)$.

In practice, however, it is difficult to analytically iterate Eq. (10.81) many times. The common practice is to replace the distribution after the i th iteration by an *optimal* distribution $f_i^\circ(g)$ that closely mimics the properties of $f_i(g)$. The optimal $f_i^\circ(g)$ is usually taken to have the following form

$$f_i^\circ(g) = [1 - R(p)]\delta(g) + R(p)\delta[g - g^\circ(p)], \quad (10.84)$$

where $g^\circ(p)$ is an *optimal* conductance. In the past, various schemes have been proposed for determining $g^\circ(p)$, one of the most accurate of which was proposed by Bernasconi (1978), according to which, if after i iterations of Eq. (10.81), $f_i(g)$ is given by

$$f_i(g) = [1 - R(p)]\delta(g) + \sum_i a_i(p)\delta(g - g_i), \quad (10.85)$$

then, $g^\circ(p)$ is approximated by

$$g^\circ(p) \simeq \exp\left[\frac{1}{R(p)} \sum_i a_i(p) \ln g_i\right]. \quad (10.86)$$

Once $g^\circ(p)$ is calculated, Eq. (10.81) is iterated again, the new distribution $f_{i+1}(g)$ and its optimal form $f_{i+1}^\circ(g)$ are determined, and so on. In practice, after a few iterations, even an initially broad $f_0(g)$ quickly converges to a stable and narrow distribution with a shape that does not change under further rescaling. The conductivity of the conductance network is simply a suitably-defined average of this distribution.

The PSRG methods are usually very accurate for 2D networks and are flexible enough to be used for anisotropic media. They have, however, two drawbacks for

3D systems. The first is that the predictions of the PSRG methods for percolation networks with *any* type of the RG cells of size $b = 2$ are not accurate. Moreover, even after the first iteration of Eq. (10.80), the renormalized conductance distribution $f_1(g)$ is very complex; if we begin with a binary distribution, $f_1(g)$ will have 73 components of the form $\delta(g - g_i)$. Hence, analytical determination of $f_2(g)$ quickly become intractable. The second drawback is that, even for a $b = 3$ RG cell, analytical determination of the RG transformation is intractable because the total number of possible configurations of the RG cell is of the order of 10^{11} . Thus, one must resort to a Monte Carlo RG method (Reynolds *et al.*, 1980), which is, however, not any simpler than direct numerical simulation of the same problem.

Young and Stinchcombe (1975) were the first to use the PSRG method for 2D percolation networks. Stinchcombe and Watson (1976) were the first to use such methods to compute the conductivity. Their work was followed by several others who proposed several variants of the PSRG methods for calculating both percolation and conduction properties of random resistor networks and other disordered media (see for example, Payandeh, 1980; Sahimi *et al.*, 1984; Sahimi, 1988a). Clearly, the PSRG methods may be used for estimating the permeability of a porous medium modeled by a network using precisely the same method that we described. One distributes the permeabilities of the bonds according to a probability density function and proceeds as above, using exactly the same formulae. Then, a renormalized permeability distribution is constructed. A Monte Carlo sampling is used to select the permeability of each cell from the joint probability distribution of the RG cells' permeabilities. The sampling and the iteration process are continued until a satisfactory representation of the permeability distribution is obtained. King (1989, 1996) used this method for calculating permeability of macroscopic porous media. Gavrilenko and Guéguen (1998) utilized it for computing the effective permeability of fractured porous media using a network model. A relatively recent review is given by Hristopulos (2003); see also Green and Paterson (2007).

10.6

Renormalized Effective-Medium Approximation

To circumvent the difficulties that the PSRG method encounters for 3D random conductance networks, Sahimi *et al.* (1983c, 1984) proposed a new method that combined the EMA and PSRG methods, and is called the renormalized EMA (REMA). To develop the method, two facts were taken advantages of:

1. Each time a conductance network is renormalized, it becomes less critical in the sense that its associated percolation correlation length ξ'_p is smaller than the original correlation length ξ_p by a factor of b (the linear size of the RG cell); see Eq. (10.79).
2. As discussed and demonstrated above, the EMA is usually accurate away from the percolation threshold if there are no long-range correlations in the porous medium.

Therefore, if one employs the EMA with the first iteration $f_1(g)$ of the original bond conductance distribution $f_0(g)$ instead of $f_0(g)$ itself, the performance of the EMA must improve. That is, in the REMA, the conductance distribution that one uses in Eq. (10.29) is $f_1(g)$, instead of $f_0(g)$. Because the bonds of the renormalized conductance network are b times longer than the original ones, this necessitates a rescaling of conductivities of the renormalized network to replicate the old one: The REMA conductivity is taken to be the same as that for the original resistor network at $p = 1$.

The REMA is significantly more accurate than *both* the EMA and PSRG. For example, the REMA predicts the bond percolation threshold p_c to be the root of the following equation

$$R(p_c) = \frac{2}{Z}, \quad (10.87)$$

which should be compared with Eq. (10.31), the prediction of the EMA. Thus, with the 3D RG cell of Figure 10.3, the RG transformation is given by

$$R(p) = p^{12} + 12p^{11}q + 66p^{10}q^2 + 220p^9q^3 + 493p^8q^4 + 776p^7q^5 + 856p^6q^6 + 616p^5q^7 + 238p^4q^8 + 48p^3q^9 + 4p^2q^{10}, \quad (10.88)$$

so that, using Eq. (10.88) in Eq. (10.87) with $Z = 6$, one obtains $p_c \simeq 0.267$, only 7% larger than the numerical estimate $p_c \simeq 0.249$ for the simple-cubic network. Moreover, so long as one uses the type of 2D self-dual RG cells that are shown in Figure 10.3, the REMA predicts the exact bond-percolation threshold of the square network for *any* cell size b .

Using the REMA, Sahimi *et al.* (1983c, 1984) and Sahimi (1988a) also obtained very accurate predictions of the effective conductivity of various 2D and 3D networks. Figure 10.4 compares the predictions of the REMA for the effective conductivity of a simple-cubic network of conductances with those of the EMA and the simulation results, and cluster theory of Ahmed and Blackman (1979). It is clear that the REMA provides very accurate predictions for the effective conductivities.

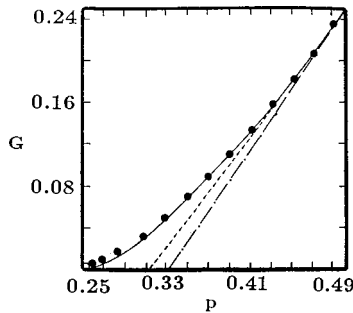


Figure 10.4 Comparison of the Monte Carlo data (circles) for the conductivity of a simple-cubic network with the predictions of the REMA (solid curve), cluster EMA of Ahmed and Blackman (1979), and the EMA (dashed-dotted curve) (after Sahimi *et al.*, 1983c).

Liu *et al.* (1992), Zhang and Seaton (1992) extended the above REMA to an arbitrary conductance distribution, and showed the extension to be highly accurate. Adler and Berkowitz (2000) compared the accuracy of the REMA, EMA, and other methods for calculating the effective conductivity of random networks. Sahimi (2003a) provides a comprehensive discussion of the subject.

10.7

The Bethe Lattice Model

None of the methods of estimating the effective permeability K_e and conductivity g_e that have been described so far are not exact and provide only approximate estimates. The only exception is the exact solution of electrical conduction in a Bethe lattice (Straley (1977), Stinchcombe (1974)). Consider a Bethe lattice of coordination number Z in which the bonds' conductance distribution is $f(g)$. It was shown (Straley, 1977) that the conductivity g_e of a Bethe lattice follows the following power law near the percolation threshold

$$g_e \sim (p - p_c)^3, \quad (10.89)$$

so that the critical exponent is $\mu_p = 3$, very different from $\mu_p \simeq 2$ for 3D systems. Thus, the conductivity of a Bethe lattice is a poor approximation to that of 3D networks. However, there is a happy coincidence here in the following sense. Suppose that a function $\Upsilon(x)$ is the solution of the following nonlinear integral equation

$$\int_0^\infty \Upsilon(x) \exp(-\lambda x) dx = \left\{ \int_0^\infty \int_0^\infty \exp\left[-\frac{gx}{\lambda(g+x)}\right] f(g) \Upsilon(x) dg dx \right\}^{Z-1}. \quad (10.90)$$

It is not possible to obtain an exact analytical solution for $\Upsilon(x)$ and, thus, Eq. (10.90) must be solved numerically. A conductivity g_m may be defined that is given by (Stinchcombe (1974))

$$g_m = Z \left[\int_0^\infty \int_0^\infty f(g) \Upsilon(x) \left(\frac{gx}{g+x} \right) dg dx \right]^{Z-2}. \quad (10.91)$$

It can be shown (Stinchcombe (1974)) that near p_c ,

$$g_m \sim (p - p_c)^2, \quad (10.92)$$

so that the critical exponent for g_m is two, identical with that of 3D networks. The difference between g_e and g_m is as follows (Straley, 1977). If we impose a unit potential gradient between any interior site of a Bethe lattice and its external boundary at infinity, then g_e is the current *per bond* that flows through the lattice. Note

that this is also the quantity that one calculates for 3D networks. Thus, g_e is the *macroscopic* conductivity of the network. On the other hand, g_m is the *total* current that flows through the lattice in the same unit potential gradient and, thus, g_m is a *microscopic* conductivity.

Heiba *et al.* (1982) proposed that g_m is an accurate approximation to the conductivity of 3D networks. The reason is as follows. g_m follows the same power law near the percolation threshold p_c as the conductivity of 3D networks. Moreover, as described in Chapter 3, the coordination number of a Bethe lattice can be adjusted so that its percolation threshold matches that of a 3D network. For example, the bond percolation threshold of the simple-cubic network is $p_c \simeq 0.249$. Thus, if we use a Bethe lattice of coordination number five, its percolation threshold would be $p_c = 1/(Z - 1) = 1/(5 - 1) = 0.25$, which is less than 0.1% larger than that of the cubic network. Thus, a Bethe lattice of coordination number five may be used for estimating the conductivity of a simple-cubic network. Figure 10.5 compares the conductivity g_e of a cubic network obtained by Monte Carlo calculations and the conductivity g_m of a Bethe lattice of coordination number five. It is evident that the difference between the two is so small that it cannot be detected with the naked eye.

Thus, g_m is an excellent estimator of the conductivity of 3D networks, *regardless* of any particular form that the conductance distribution $f(g)$ may have. The same method may be used for estimating the permeability of 3D networks. Sahimi (1988b) extended this concept to the case of unsteady-state diffusion and a first-order chemical reaction, while Sahimi (1993a) extended the analysis to the case in which a non-Newtonian fluid flows through a disordered porous medium represented by a pore network. Sahimi (2003a) described the application of the same concept to estimating other important transport properties.

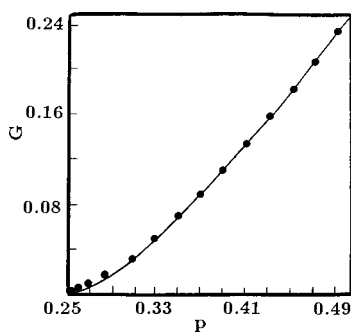


Figure 10.5 Comparison of the conductivity of a simple-cubic network computed by the Monte Carlo method (circles) with the predictions of a Bethe lattice of coordination number five.

10.8

Critical Path Analysis

A powerful idea for analyzing transport in heterogeneous media is the *critical path* analysis (CPA). The concept was first proposed by Ambegaokar, Halperin and Langer (AHL) in 1971. They argued that transport in a disordered medium with a *broad* distribution of conductances is dominated by those with magnitudes that are larger than some characteristic conductance g_c . The characteristic conductance g_c is defined as the smallest conductance such that the set of conductances $\{g | g > g_c\}$ forms a conducting sample-spanning cluster. This cluster is called the critical path. Therefore, transport in a disordered medium with a broad conductance distribution reduces to a percolation problem with threshold conductance g_c . Shante (1977) and Kirkpatrick (1979) modified the critical path concept by assigning g_c to all the local conductances g with $g \geq g_c$, and setting all the conductances g with $g < g_c$ to zero (since the contribution of such conductances to the overall conductivity is very small). They then arrived at a trial solution for the sample conductance of the form

$$g_e = a g_c [p(g_c) - p_c]^{\mu_p}, \quad (10.93)$$

which is just the power law for the conductivity of the network described in Chapter 3. Here, $p(g_c)$ denotes the probability that a given conductance is greater than or equal to g_c , and a is a constant. Equation (10.93) is then maximized with respect to g_c to obtain an estimate of it and, thus, an estimate for g_e . Computer simulations of Berman *et al.* (1986) for 2D networks with various conductance distributions (Gaussian, log-normal, and uniform) confirmed the quantitative accuracy of the AHL idea, even for relatively narrow conductance distributions. Therefore, calculating the effective transport properties of disordered media in which percolation may not seem to play any role at all may be reduced to determining the same properties for a percolation network if the statistical distribution of the local flow or transport properties is broad. Hunt and Ewing (2009) describe many applications of the CPA to various problems in porous media.

Katz and Thompson (1986, 1987) extended the critical path method to estimate the permeability and electrical conductivity of (fluid-saturated) porous media. In a porous medium, the local hydraulic conductance is a function of a length scale l . Therefore, the critical conductance g_c defines a corresponding characteristic length scale l_c . Since both flow and electrical conduction problems belong to the same class of percolation problems, the length scale that signals the percolation threshold in the flow problem also defines the threshold in the electrical conductivity problem. Thus, we rewrite Eq. (10.93) as

$$g_e = \phi g_c(l) [p(l) - p_c]^{\mu_p}, \quad (10.94)$$

where the porosity ϕ ensures a proper normalization of the fluid or the electric-charge density. The function $g_c(l)$ is equal to $c_f l^3$ for the flow problem and to $c_e l$ for the conduction problem. For appropriate choices of the function $p(l)$, the conductance $g_e(l)$ achieves a maximum for some $l_{\max} \leq l_c$. In general, the length scale

$l_{\max}^f$ for the flow problem is different from $l_{\max}^c$ for the conduction problem because the flow and transport paths have different weights in the two problems.

If $p(l)$ allows for a maximum in the conductance that occurs for $l_{\max} \leq l_c$, then we may write

$$l_{\max}^f = l_c - \Delta l_f = l_c \left[1 - \frac{\mu_p}{1 + \mu_p + l_c \mu_p \frac{p''(l_c)}{p'(l_c)}} \right], \quad (10.95)$$

$$l_{\max}^c = l_c - \Delta l_c = l_c \left[1 - \frac{\mu_p}{3 + \mu_p + l_c \mu_p \frac{p''(l_c)}{p'(l_c)}} \right]. \quad (10.96)$$

If the pore-size distribution of the porous medium is very broad, then $l_c \mu_p p''(l_c)/p'(l_c) \ll 1$, and Eqs. (10.95) and (10.96) reduce to

$$l_{\max}^f = l_c \left(1 - \frac{\mu_p}{1 + \mu_p} \right) \simeq \frac{1}{3} l_c, \quad (10.97)$$

$$l_{\max}^c = l_c \left(1 - \frac{\mu_p}{3 + \mu_p} \right) \simeq \frac{3}{5} l_c, \quad (10.98)$$

if μ_p , the exponent for the power-law behavior of the effective conductivity and permeability near the percolation threshold, is taken to be two. We can then establish a relation between g_e and K_e . Writing

$$g_e = a_1 \phi [p(l_{\max}^c) - p_c]^{\mu_p}, \quad (10.99)$$

and

$$K_e = a_2 \phi \left(l_{\max}^f \right)^2 [p(l_{\max}^f) - p_c]^{\mu_p}, \quad (10.100)$$

we obtain to first order in Δl_c or in Δl_f ,

$$p(l_{\max}^{f,c}) - p_c = -\Delta l_{f,c} p'(l_c). \quad (10.101)$$

To obtain the constants a_1 and a_2 , Katz and Thompson (1986) assumed that at the local level, the conductivity of a porous medium is g_f – the conductivity of the fluid (usually brine) that saturates the pore space – and that the local pore geometry is cylindrical. These assumptions imply that $a_1 = g_f$ and $a_2 = 1/32$. Therefore, one obtains

$$K_e = a_3 l_c^2 \frac{g_e}{g_f}, \quad (10.102)$$

where $a_3 = 1/226$. A similar argument leads to (Katz and Thompson, 1987)

$$\frac{g_e}{g_f} = \frac{l_{\max}^c}{l_c} \phi S(l_{\max}^c), \quad (10.103)$$

where $S(l_{\max}^c)$ is the volume fraction (saturation) of the connected pore space with pore widths of size $l_{\max}^c$ and larger.

Equations (10.102) and (10.103) involve no adjustable parameters. Every parameter is fixed and precisely defined. To obtain the characteristic length l_c , Katz and Thompson (1986, 1987) proposed the use of mercury porosimetry as described in Chapter 4. As we discussed there, mercury porosimetry is a percolation process. Hence, consider a typical mercury porosimetry curve in which the pore volume of the injected mercury is measured as a function of the pressure (see Figure 10.6). In the initial portion of the curve – the part before a sample-spanning cluster of pores filled with mercury has been formed – the curvature is positive. Then, there is also an inflection point beyond which the pore volume increases rapidly with the pressure. The inflection point signals the formation of the sample-spanning cluster. Therefore, from the Washburn equation (Chapter 4), we must have $l \geq -4\sigma_{mv} \cos \theta / P_i$, where P_i is the pressure at the inflection point, σ_{mv} is the surface tension between the mercury and the vacuum, and θ is the contact angle between the mercury and the pore surface. Hence,

$$l_c = -\frac{4\sigma_{mv} \cos \theta}{P_i} \quad (10.104)$$

defines the characteristic length l_c .

Figure 10.7 compares the logarithm of the permeability for a set of sandstones calculated using Eq. (10.102) with the data. The dashed lines mark a factor of two. No adjustable parameter was used, and the agreement between the predictions and the data is very good. Note that once l_c is determined from a mercury injection curve, the saturation $S(l_{\max}^c)$ may also be determined immediately. Figure 10.8 compares the calculated and measured effective (normalized) conductivity g_e/g_f . As in the case of the permeability, no adjustable parameter was used. Katz and Thompson (1986, 1987) contend that l_c can be estimated from mercury injection curves with an error of at most 15%. The error in the constant $1/226$ in Eq. (10.102) may, however, be as large as a factor of two. We emphasize that Eqs. (10.102) and (10.103) are *not* exact, but provide very accurate estimates of K_e and g_e . Note that if, as discussed in Chapter 3, the distribution of the local conductances or permeabilities of

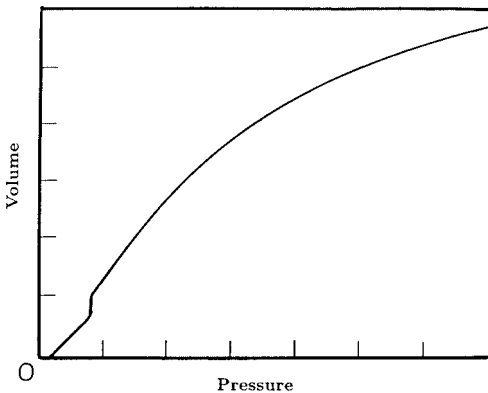


Figure 10.6 Schematic volume–pressure curve during mercury porosimetry.

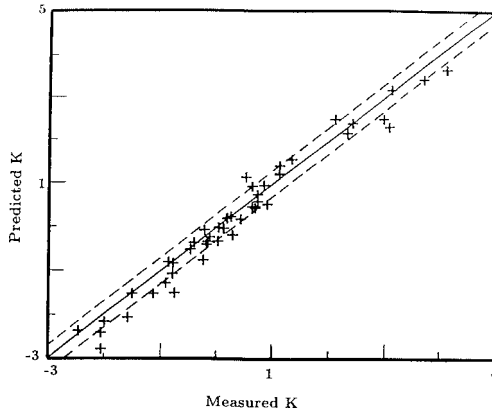


Figure 10.7 Logarithmic comparison of the predicted permeability K_e (in millidarcy) with the measured K_e (after Katz and Thompson, 1986).

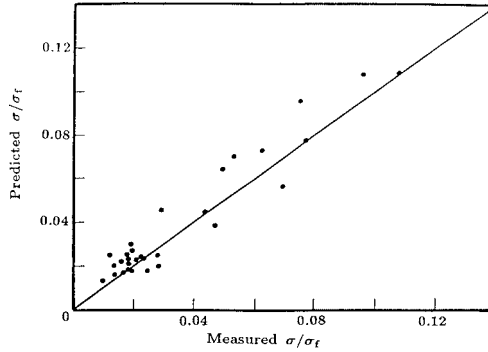


Figure 10.8 Logarithmic comparison of the predicted conductance g_e with the data. g_f is the conductivity of the fluid saturating the porous medium (after Katz and Thompson, 1987).

a porous medium is very broad, the exponent μ_p may be nonuniversal and, therefore, its nonuniversal estimate should be used in Eqs. (10.97) and (10.98).

Some related relevant works should be mentioned here. Swanson (1981) had already recognized that during mercury injection (or flow of any non-wetting fluid in a porous medium), large pores dominate the flow paths, and that the inflection point in the pore volume–pressure curve signals the formation of a sample-spanning cluster. Thus, he postulated a relation between the effective permeability and the capillary pressure curve. He maximized the product of the length and saturation, both of which can be estimated from the mercury-injection curve, and obtained

$$l \sim (S^2 l^2)_{\max}, \quad (10.105)$$

where $l_{\max}$ is a length scale very similar, both qualitatively and numerically, to the length scale l_c introduced by Katz and Thompson (1986, 1987). The agreement

between the predictions of Eq. (10.105) and the data was very good, which is not entirely surprising given the similarity between Swanson's $l_{\max}$ (not to be confused with Katz and Thompson's $l_{\max}$) and l_c . It should, however, be noted that the transport paths considered by Swanson were appropriate for electrical conduction, but not for the fluid flow problem. Thus, the agreement could be fortuitous.

Banavar and Johnson (1987) and Le Doussal (1989) calculated the coefficient a_3 in Eq. (10.102) slightly differently than the way Katz and Thompson (1986, 1987) did. For example, Banavar and Johnson (1987) estimated that $a_3 \simeq 7.68 \times 10^{-3}$, which should be compared with Katz and Thompson's estimate, $a_3 = 1/226 \simeq 4.42 \times 10^{-3}$. Banavar and Johnson's predictions are still within the error bars of Figure 10.8. Nyame and Ilbston (1980) used an empirical parameter similar to l_c to describe the permeability of cement paste. Hagiwara (1984) replaced S^2 in Eq. (10.103) with g_e to obtain $K_e \sim g_e l^2$, which appears again to agree with data.

Friedman and Seaton (1998) carried out an extensive study of the effective permeability and electrical conductivity of 3D pore networks using numerical simulations and the critical path analysis. Hunt and Gee (2002) used the critical path method to study fractal porous media, and compared the results with the data from the Hanford site (in the Washington state). Hunt (2005) utilized continuum percolation (Chapter 3) and the critical path method to study saturation-dependence of air permeability.

The critical path method may also be extended in order to calculate the permeability of fractured rocks. Indeed, Charlaix *et al.* (1987a) used arguments very similar to those of Katz and Thompson to calculate the permeability of fracture networks with a broad distribution of fracture apertures. Finally, the critical path method may also be used for estimating the permeability of a porous medium saturated by a *non-Newtonian* fluid (Sahimi, 1993a).

10.9

Random Walk Method

As discussed in Section 10.2.3 (as well as in Chapter 9), from the time dependence of the mean-square displacement of a random walker, the effective diffusivity and, hence, the effective conductivity g_e , of porous media may be calculated. In the context of random conductance networks, Brandt (1975) was the first to have used the method, but it was de Gennes (1976) who popularized the method by making an analogy between the motion of a random walker in a disordered medium and that of an ant in a labyrinth. Mitescu and Rousseny (1976) followed de Gennes' idea and performed relatively extensive simulations on percolation clusters. The developments in this area were reviewed extensively by Hughes (1995).

In Chapter 9, we described how the method may be used to simulate diffusion, conduction, and flow in porous media represented by packings of overlapping or non-overlapping particles. In the context of the random conductance networks, Bunde *et al.* (1985) considered a general two-component network in which the conductivity of the bonds was either g_a or g_b , and formulated a random walk model

for calculating the effective conductivity of the network. In their model, one first fixes a time scale by noting that $g_a \sim D_A \sim \ell^2/t_A$, where D_A is the diffusivity in the region A, ℓ is the mean-free path (or the distance traveled by a random walker), and τ_A is the characteristic time for traveling a distance ℓ . Therefore,

$$\frac{\tau_A}{\tau_B} = \frac{g_a}{g_B}. \quad (10.106)$$

The transition probability W_i , the probability that the random walker takes a step to its i th neighbor. This probability is given by

$$W_i = \frac{p_i}{\sum_i p_i}, \quad (10.107)$$

where $p_i = g_a$ or g_B . More generally, if each bond $i j$ of a network has a conductance g_{ij} , then the transition probability W_{ij} for a step from i to j is given by

$$W_{ij} = \frac{g_{ij}}{\sum_j g_{ij}}. \quad (10.108)$$

Therefore, one may use a random walk method to simulate diffusion in a porous medium with a continuous (or discrete) distribution of local conductances. The effective conductivity of the system will then be exactly equal to its effective diffusivity.

In practice, one must use certain tricks in order to speed up the simulations. For example, if $g_a \ll g_B$, then the random walker may spend a long time in the A-region before making a transition to the B-region, and vice versa. Using vectorization (for use in a vector or supercomputer), a highly efficient computer algorithm for simulating random walks in a disordered medium, was also developed (Sahimi and Stauffer, 1991). For more recent applications of this method to composite materials see van Sichen (2002b).

10.10

Non-Darcy Flow

As described in Chapter 9, if the velocity of a fluid flowing in a porous medium is high enough, Darcy's law breaks down. In that case, additional terms are added to the basic Darcy's law, for example, that in the Forchheimer equation, Eq. (9.163), which adds a terms that is proportional to the square of the flow velocity. Pore network models have been used in the past to study non-Darcy flow in porous media, and have helped to gain a better understanding of the conditions under which such flows better describe fluid flow in porous media.

Thauvin and Mohanty (1998) developed a pore-network model to study high velocity fluid flow in a porous medium. In their model, in addition to the pore throats,

pore bodies were also assigned effective sizes. Recognizing that in a porous medium the interstitial stream lines are not straight lines, but rather converge and diverge as they travel from pore bodies to pore throats, and that they bend at fluid junctions because the pore throat hydraulic conductances are distributed, they included, in the pore-level flow equations, additional terms that account for the pressure drops due to such bending of the stream lines and convergent-divergent segments of the pore body-pore throat junctions. Thus, the additional pressure loss due to the bending of flow at a pore body was approximated by

$$\Delta P_b = 0.9 f \rho V^2, \quad (10.109)$$

where f is the fraction of the fluid from each pore throat that bends, which can be calculated by knowing flow rates in all the incoming and outgoing pore throats (at a given pore body), assuming perfect mixing at each pore body. The pressure drop due to the expansion part of the pore body-pore throat junction was taken to be

$$\Delta P_e = \frac{1}{2} \rho V^2 \left[1 - \left(\frac{R_t}{R_b} \right)^2 \right] \left(\frac{R_t}{R_b} \right)^2, \quad (10.110)$$

whereas the pressure drop due to the contraction part of the junction was taken to be

$$\Delta P_c = \frac{1}{2} \rho V^2 \left[1.45 - 0.45 \left(\frac{R_t}{R_b} \right)^2 - \left(\frac{R_t}{R_b} \right)^4 \right]. \quad (10.111)$$

Assuming that the pore throats are cylindrical, the usual (Darcy) pressure drop between the centers of two adjacent pore bodies due to viscous forces is given by the usual equation,

$$\Delta P_d = \frac{8 \mu l_t}{R_t^2} V, \quad (10.112)$$

where l_t is the pore throat's length. Then, the total pressure drop between two adjacent pore bodies is the sum of the four pressure drops.

One important result of the pore network simulations of Thauvin and Mohanty (1998) was establishing a correlation between the parameter β of the Forchheimer equation defined by Eq. (9.163), and the characteristics of the pore network. The correlation proposed is given by

$$\beta = 1.55 \times 10^4 \tau^{3.35} K_e^{-0.98} \phi^{-0.29}, \quad (10.113)$$

where τ is the tortuosity of the pore network. Equation (10.113) should be compared with those presented in Section 9.13, and in particular, with Eq. (9.167). Non-Darcy flow in anisotropic porous media was also studied by Wang *et al.* (1999), using a pore network model. They developed an extension of the Forchheimer equation in tensorial form, suitable for anisotropic porous media. Lao *et al.* (2004) also developed a pore network model to study non-Darcy flow in porous media.

11

Dispersion in Flow through Porous Media

Introduction

In Chapters 9 and 10, we described and discussed flow and transport processes that involve only one fluid and one fluid phase. Beginning with this chapter, we take up the flow phenomena that are at the next level of complexity, namely, those that involve at least two fluids and one or more fluid phases. One of the most important of such phenomena is hydrodynamic dispersion that involves two fluids, and is described and studied in the present chapter. Miscible displacements, which represent generalizations of the dispersion processes (in which the viscosities of the two fluids are *not* equal) will be studied in Chapter 13, while multiphase flows will be described in Chapters 14 and 15.

11.1

The Phenomenon of Dispersion

When two miscible fluids are brought into contact with an initially sharp front separating them, a transition zone develops across the initial front, the two fluids slowly diffuse into one another, and after some time, a diffused mixed zone develops. If one assumes that the volumes of the two fluids do not change upon the mixing, then the net transport of one of the solute across any arbitrary plane is represented by the Fick's second law of diffusion

$$\frac{\partial C}{\partial t} = D_m \nabla^2 C . \quad (11.1)$$

Here, C is solute the concentration, t is the time, and D_m is the molecular diffusivity of the solute in the solvent. The mixing of the two fluids is independent of whether or not there is a convective current through the medium. If, however, the solvent is also flowing, then there will be some additional mixing of a different sort – *convective mixing* – which is caused by a nonuniform velocity field that, in turn, is caused by the morphology of the medium, the fluid flow condition (fast or slow) and the chemical or physical interactions with the solid surface of the medi-

um. The resulting mixing process is called *hydrodynamic dispersion*, or dispersion for the sake of brevity.

Dispersion is important to a wide variety of processes, for example, miscible displacements in enhanced oil recovery, salt water intrusion in coastal aquifers where fresh and salt waters mix by dispersion, and in-situ study of the characteristics of aquifers, where a classical method of determining the characteristics is injecting fluid tracers into the aquifers and measuring their travel times. In addition, pollution of the surface waters due to industrial and nuclear wastes is another important problem in which dispersion plays a fundamental role. In particular, burying nuclear wastes deep in the rock has been suggested as a way of sequestering the pollution that they cause, but leakage of the wastes into groundwater aquifers has been a major concern. Dispersion also occurs in flow and reaction in packed-bed chemical reactors and has been studied extensively by chemical engineers for a long time; see, for example, Bernard and Wilhelm (1950)¹⁾ as one of the earliest papers on the subject.

11.2

Mechanisms of Dispersion Processes

In steady flow through a disordered porous medium, the *transit time*, or *first-passage time*, of a fluid particle or tracer between the entrance and exit planes depends on the path, or streamline that it follows through the pore space. A population of tracers passing the entrance plane at the same instant will arrive at the exit plane by a set of streamlines with a *distribution of transit times*. Thus, a solute concentration front will spread in the mean-flow direction as it passes through the medium. The resulting first-passage time distribution (FPTD) is a measure of *longitudinal dispersion* in a porous medium.

Likewise, a population of tracers or particles passing simultaneously through a restricted area of the entrance plane will not entirely follow the mean flow to the exit plane, but will be dispersed in the transverse directions (perpendicular to the direction of the mean flow) as well. That is, the population and the set of streamlines traveled will have a wider distribution of exit locations than of the entrance ones. Thus, a concentration front will also spread laterally on the way to the exit plane. The distribution of the first-passage times for crossing the porous medium at a given transverse plane is a measure of *transverse dispersion* in the porous medium.

Two basic mechanisms drive dispersion in macroscopically homogeneous, microscopically disordered porous media, and arise in the pore-level velocity field forced on the flowing fluid by the irregularity of the pore space.

1) *Richard Herman Wilhelm* (1909–1968), Chairman of Chemical Engineering Department at Princeton University, made significant contributions to fixed-bed catalytic reactors, fluidized transport and separation processes. The Wilhelm Award of the American Institute of Chemical Engineers has been named in his honor.

1. The first mechanism is *kinematic*: streamtubes divide and rejoin repeatedly at the junctions of flow passages in the highly interconnected pore space. The consequent tangling and divergence of streamlines is accentuated by the widely varying orientations of flow passages and coordination numbers of the pore space. The result is a wide variation in the lengths of the streamlines and their downstream transverse separations.
2. The second mechanism is *dynamic*: the speed with which a given flow passage is traversed depends on the flow resistance or hydraulic conductance of the passage, its orientation, and the local pressure field.

The two mechanisms conspire to produce broad FPTDs between the entrance and exit planes. The two mechanisms also suggest two possible geometrical aspects of the dispersion processes defined with respect to the mean-velocity direction, that is, a longitudinal effect due to the difference between the velocity components in the direction of mean flow, and a transverse effect due to the differences between *local* velocity components orthogonal to the direction of the mean flow.

The two mechanisms of dispersion do *not* depend on molecular diffusion. Diffusion modifies, however, the effects of the two basic mechanisms by moving material from one streamline to another as well as by the usually weaker streamwise diffusion of material relative to the average velocity. The solid matrix of a porous medium acts locally, of course, as a separator of the streamlines and, thus, as a barrier to diffusion. Therefore, the modification of dispersion by diffusion depends on the pore space morphology and how it affects local flow and concentration fields. The effect of molecular diffusion is usually important only at the pore level, where it acts to transfer the tracer particles out of slow or stagnant regions of the pore space.

11.3

The Convective-Diffusion Equation

Dispersion processes in microscopically disordered and macroscopically isotropic and homogeneous porous media are usually modeled based on the convective-diffusion (CE) equation:

$$\frac{\partial C}{\partial t} + \langle \mathbf{v} \rangle \cdot \nabla C = D_L \frac{\partial^2 C}{\partial x^2} + D_T \nabla_T^2 C, \quad (11.2)$$

where $\langle \mathbf{v} \rangle$ is the macroscopic mean velocity, C is the mean concentration of the solute, and ∇_T^2 is the Laplacian in the transverse directions. For the sake of simplicity, we delete $\langle \cdot \rangle$ and denote the magnitude of the average fluid velocity vector by V or V_m . Thus, the basic idea is to model dispersion processes as *anisotropic* diffusional spreading of the concentration, with the diffusivities being the longitudinal dispersion coefficient D_L and the transverse dispersion coefficient D_T . One important goal of any study of dispersion in porous media is to investigate the conditions under which it *cannot* be represented by the CD equation.

Dispersion is said to be *diffusive* or *Gaussian* if it is described by the CD equation. If a population of the solute particles is injected into the medium at $\mathbf{r}_0 = (x_0, y_0, z_0)$ at $t = 0$ (i.e., $C(x_0, y_0, z_0, t) = C_0 \delta(t)$), for diffusive dispersion, the probability density $P(\mathbf{r}, t)$ follows the Gaussian distribution

$$P(\mathbf{r}, t) = (8\pi^3 D_L D_T^2 t)^{-\frac{3}{2}} \exp \left[-\frac{(x - x_0 - Vt)^2}{4D_L t} - \frac{(y - y_0)^2}{4D_T t} - \frac{(z - z_0)^2}{4D_T t} \right], \quad (11.3)$$

where $P(\mathbf{r}, t)d\mathbf{r}$ is the probability that a solute particle is in a plane between $\mathbf{r}$ and $\mathbf{r} + d\mathbf{r}$ at time t , and $\mathbf{r} = (x, y, z)$. $P(\mathbf{r}, t)$ is proportional to C/C_0 and, therefore, Eq. (11.3) represents a solution of Eq. (11.2). If one defines $\mathcal{Q}(\zeta - \zeta_0, t)dt$ as the probability that a solute particle, beginning in the plane at ζ_0 will cross, for the first time, a plane at ζ between t and $t + dt$. Then, from Eq. (11.3), one can easily obtain the FPTD in a given direction since $\mathcal{Q}$ and P are related:

$$P(\zeta - \zeta_0, t) = \int_0^t P(\zeta - \zeta_1, t - \tau) \mathcal{Q}(\zeta_1 - \zeta_0, \tau) d\tau, \quad (11.4)$$

and, therefore

$$\mathcal{Q}(\zeta - \zeta_0, t) = |\zeta - \zeta_0| (4\pi D_\zeta t^3)^{-\frac{1}{2}} \exp \left[-\frac{(\zeta - \zeta_0 - V_\zeta t)^2}{4D_\zeta t} \right], \quad (11.5)$$

where D_ζ and v_ζ are the dispersion coefficient and the mean flow velocity in the ζ -direction, respectively. The moments of $\mathcal{Q}$ yield information about the flow field and the dispersion processes. For example, the first two moments of the FPTD in the longitudinal direction are given by

$$\langle t \rangle = \frac{L}{V}, \quad (11.6)$$

and

$$\langle t^2 \rangle = \langle t \rangle^2 \left(1 + \frac{2D_L}{LV} \right), \quad (11.7)$$

where $L = \zeta - \zeta_0$. In general, one can easily show that for large L and to the leading order, one has

$$\langle t^n \rangle \sim \langle t \rangle^n, \quad (11.8)$$

where $n > 1$ is an integer. Equation (11.8) holds true as long as the description of dispersion by the CD equation is valid. Therefore, one way of showing that a CD equation cannot describe the dispersion process in a given porous medium is to show that $\langle t^n \rangle / \langle t \rangle^n$ ($n > 1$) is *not* a constant (i.e., the ratio depends on t) and one requires more information than just $\langle t \rangle$ to describe the moments of the FPTD. This issue will be discussed later in this chapter.

11.4

The Dispersivity Tensor

Because dispersion is an anisotropic phenomenon, and since one must have invariance under coordinate transformations, the dispersion coefficients constitute a tensor. One may then ask, what is the most general form of the tensor? This question was addressed by Bear (1961) and Scheidegger (1961). In particular, Bear (1961) noted that the dispersion coefficients should be written as

$$D_{ik} = a_{iklm} \frac{v_l v_m}{|V|}, \quad (11.9)$$

where $|V|$ is the magnitude of the fluid velocity. The coefficient a_{iklm} is called the *dispersivity*, and represents the typical length scale over which significant dispersion occurs. Clearly, a_{iklm} is a fourth-rank tensor with 81 components. However, several symmetry properties significantly reduce the number of independent components of the tensor. One is that $a_{iklm} = a_{ikml}$. That this symmetry should hold is obvious. If the equality did not hold, the two coefficients could be made to be because that would not affect the general form of the equation of motion. Due to Onsager's principle of microscopic reversibility, one must also have $a_{iklm} = a_{kilm}$. The two symmetries reduce the number of independent components of the dispersivity tensor $\mathbf{a}$ from 81 to 36.

To simplify the notation, it is customary to write a tensor with four indices and only 36 nonzero components in the form of a 6×6 matrix. Suppose that we denote the matrix by $a_{\alpha\beta}$. The connection between the Latin and Greek notations is as follows. To every pair (x_i, x_j) (for $i, j = 1, 2$, and 3), we attribute a corresponding value of α . Let $x_1 = x$, $x_2 = y$, and $x_3 = z$. Then, $\alpha = 1, 2, 3, 4, 5, 6$ correspond to (x, x) , (y, y) , (z, z) , (x, y) , (x, z) , (y, z) , and similarly for β . Scheidegger (1961) showed that if we require invariance for 90° rotations around the x and z axes, the tensor $[a_{\alpha\beta}]$ reduces to

$$[a_{\alpha\beta}] = \begin{bmatrix} a_{11} & a_{12} & a_{12} & 0 & 0 & 0 \\ a_{12} & a_{11} & a_{12} & 0 & 0 & 0 \\ a_{12} & a_{12} & a_{11} & 0 & 0 & 0 \\ 0 & 0 & 0 & a_{44} & 0 & 0 \\ 0 & 0 & 0 & 0 & a_{44} & 0 \\ 0 & 0 & 0 & 0 & 0 & a_{44} \end{bmatrix}$$

so that $[a_{\alpha\beta}]$ is completely symmetric and has only three independent components. For an isotropic porous medium, one can also show that $a_{44} = \frac{1}{2}(a_{11} - a_{12})$, so that the dispersivity tensor only has two independent components and, therefore, there are only two dispersion coefficients, the longitudinal and transverse dispersion coefficients, D_L and D_T , and the corresponding dispersivities, a_L and a_T . More recently, Salles *et al.* (1993) studied dispersion in porous media in order to determine the dispersion tensor.

11.5

Measurement of the Dispersion Coefficients

Since measurement of D_L and D_T is not as straightforward as that of the effective permeability K_e or the effective diffusivity D_e , we describe the measurement techniques. In principle, there are two distinct ways of observing and measuring the dispersion phenomenon in a porous medium. One may observe the variations of the solute concentration either as a function of the distance from the entrance to the medium *at a fixed time*, or as a function of the time *at a fixed distance* from the entrance.

11.5.1

Longitudinal Dispersion Coefficient

The longitudinal dispersion coefficient D_L can be measured by at least three methods that are all based on the concentration profile of the solute. However, while in one method the profile is measured directly, the profile is inferred from the measurement of other quantities in the other two methods.

11.5.1.1 Concentration Measurements

Laboratory measurements of D_L are usually carried out in porous media that are effectively one dimensional under a constant fluid velocity v . The initial and boundary conditions are: $C(x \geq 0, t = 0) = 0$, $C(x = 0, t > 0) = C_0$, and $C(x \rightarrow \infty, t \geq 0) = 0$. Then, the solution to the one-dimensional (1D) version of Eq. (11.2) is obtained by the Laplace transform technique. Assuming that the length of the porous medium is L , and defining the dimensionless parameters

$$\alpha_{\pm} = \frac{x \pm Vt}{(4D_L t)^{\frac{1}{2}}} , \quad (11.10)$$

the solution of the CD equation, subject to the aforementioned initial and boundary conditions, is given by

$$\frac{C}{C_0} = \frac{1}{2} \operatorname{erfc}(\alpha_-) + \frac{1}{2} \exp\left(\frac{xV}{D_L}\right) \operatorname{erfc}(\alpha_+) , \quad (11.11)$$

where $\operatorname{erfc}(z)$ is the complementary error function. Compared with the first term, the second term of Eq. (11.11) is usually very small and, therefore, can be neglected without any significant error, in which case

$$\frac{C}{C_0} = \frac{1}{2} \operatorname{erfc}(\alpha_-) = \frac{1}{\sqrt{\pi}} \int_{\alpha_-}^{\infty} \exp(-\kappa^2) d\kappa = \frac{1}{2} [1 - \operatorname{erf}(\alpha_-)] . \quad (11.12)$$

Equation (11.11) is rewritten as

$$\frac{C}{C_0} = \frac{1}{\sqrt{2\pi}} \int_{\alpha'}^{\infty} \exp\left(-\frac{\kappa^2}{2}\right) d\kappa , \quad (11.13)$$

where $\alpha' = 2\alpha_-/\sqrt{2}$. Figure 11.1 shows the concentration profiles C/C_0 versus $x_d = x/L$ and $t_d = \int_0^t V dt/(L\phi)$ (a dimensionless time), where ϕ is the porosity of the porous medium. According to Eq. (11.12), at a fixed time, the solution is a normal or Gaussian distribution function $1 - N[(x - \langle x \rangle)/s]$ with the average $\langle x \rangle = Vt$ and the standard deviation $\sigma = \sqrt{2D_L t}$. Two well-known properties of a normal distribution are

$$N(1) \simeq 0.84, \quad N(-1) \simeq 0.16, \quad (11.14)$$

which allow us to measure D_L . The width w of the transition or mixing zone, that is, the zone between a region with pure solute and one with pure solvent, is usually defined as the difference between the values of x at which $C/C_0 = 0.16$ and $C/C_0 = 0.84$. Thus,

$$w = 2\sigma = 2\sqrt{2D_L t} = x_{0.16} - x_{0.84}, \quad (11.15)$$

from which we obtain

$$D_L = \frac{(x_{0.16} - x_{0.84})^2}{8t}. \quad (11.16)$$

Thus, if we construct a graph of C versus x at a fixed time t , we can determine D_L based on Eq. (11.16). Alternatively, we can fit Eq. (11.13) to the data for the concentration, with D_L being the fitting parameter.

In many cases, it is easier to fix x (for example, at the exit of the porous medium) and measure the concentration as a function of time t . If so, Eqs. (11.13) and (11.14) provide an expression for D_L :

$$D_L = \frac{1}{8} \left(\frac{x - Vt_{0.16}}{\sqrt{t_{0.16}}} - \frac{x - Vt_{0.84}}{\sqrt{t_{0.84}}} \right)^2, \quad (11.17)$$

where $t_{0.16}$ is the time at which $C/C_0 = 0.16$.

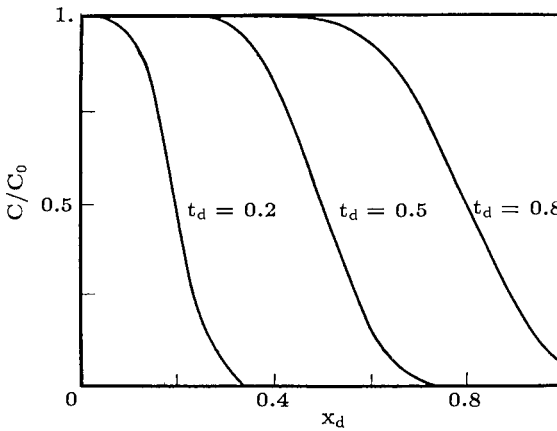


Figure 11.1 Normalized concentration profile C/C_0 versus the dimensionless distance x_d at various dimensionless times t_d .

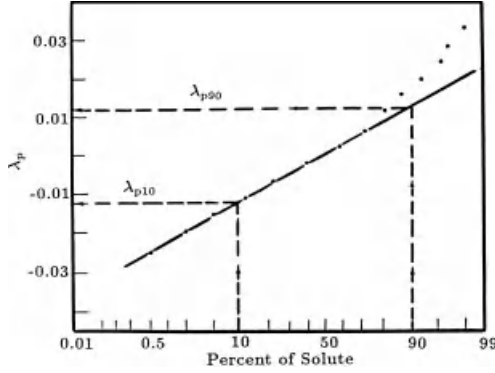


Figure 11.2 A typical solute-composition plot for determining the dispersion coefficient D_L on the arithmetic-probability paper. Circles represent the data (after Brigham *et al.*, 1961).

Measurement of D_L in flow through unconsolidated porous medium is usually easier than that for consolidated ones. Thus, we describe methods for measuring D_L (and D_T) in such porous media, although similar methods may also be used for consolidated porous media. One saturates a packed column with one fluid (the solvent), displaces it with another miscible fluid (the solute), and measures the fluid composition at the exit end of the column as a function of the displacement. Brigham *et al.* (1961) developed a convenient method for estimating D_L from data of this type. In this method, one plots $\lambda_p = (v/V_c - 1)/\sqrt{v/V_c}$ versus the percent of the solute on an arithmetic probability paper, where v is the volume of the solute injected into the porous medium, and V_c is the column's volume. Then, D_L is given by

$$D_L = vL \left(\frac{\lambda_{p90} - \lambda_{p10}}{3.625} \right)^2, \quad (11.18)$$

where L is the length of the column, and λ_{p90} is the value of λ_p when the solvent contains 90% displacing fluid. Equation (11.18) is derived by an argument similar to that for deriving Eq. (11.16), except that the mixing (transition) zone is defined as the region between the axial positions x at which $C/C_0 = 0.1$ and 0.9 . Figure 11.2 shows the schematic representation of the method.

11.5.1.2 Resistivity Measurements

In this method (see, for example, Odling *et al.*, 2007; Aggelopoulos and Tsakiroglou, 2007, for recent references), one measures the resistivity of the porous medium that is filled by an electrically-conductive fluid (the solvent) together with the injected fluid (the solute). However, the DC resistivity (or conductivity) cannot be measured because the ion drift toward each electrode generates a blocking counter voltage. Therefore, the AC measurements are carried out. The typical procedure is as follows (Odling *et al.*, 2007).

The sample is first saturated with low-salinity brine and its impedance is measured. Then, a very slow reverse flow of the low-salinity brine is carried out in order to block any possible upflow into the sample. This is followed by injection from the bottom of the sample of a high-salinity brine at high speed, so that the composition of fluid at the inflow end of the core is nearly changed instantaneously. The flow rate of the high-salinity solution is then reduced to what is needed for the actual dispersion experiment. The bottom piston drain of the system is then closed, the upper piston drain is opened, hence letting the high-salinity brine to be convected across the porous sample. During the entire time, a large number of impedance measurements are also carried out. The impedance decreases as the high-salinity solution displaces and expels the low-salinity solution. When the impedance no longer changes, the experiment is terminated. In addition, the total fluid flux is monitored and measured.

At any point x along the sample, the resistance $\mathcal{R}(x)$ of the porous medium is inversely proportional to the fluid saline concentration $C(x)$. Since each segment of the sample has a resistance, and the sample is effectively 1D, the effective resistance $\mathcal{R}_e$ of the sample is the sum of all the individual resistances $\mathcal{R}$. Therefore,

$$\mathcal{R}_e = c \int_0^L \frac{1}{C(x)} dx, \quad (11.19)$$

where c is the proportionality factor. If dispersion is Gaussian, which is expected to be as a laboratory-scale porous sample rarely gives rise to a non-Gaussian dispersion, then the concentration profile is given by Eq. (11.11). Thus,

$$\mathcal{R}_e = c \int_0^L \frac{2}{C_0} \frac{1}{\operatorname{erfc}(\alpha_-) + \exp\left(\frac{xV}{D_L}\right) \operatorname{erfc}(\alpha_+)} dx, \quad (11.20)$$

where $\alpha_{\pm}$ is given by Eq. (11.10). Thus, the resistance-time response of the porous sample should be fitted to Eq. (11.20), with the fitting parameters being the proportionality factor c and the dispersion coefficient D_L . As such, the problem is highly nonlinear, and careful numerical integration must be used. As an initial guess for D_L , one can use the fact that D_L is always larger than the effective molecular diffusivity D_e in the same porous sample. Though somewhat tedious, the method has proven to be highly accurate.

11.5.1.3 The Acoustic Method

In an acoustic technique, the concentration profile $C(x, t)$ is determined by measuring the variations of the velocity of a sound wave at several cross sections of the sample (Bacri *et al.*, 1984, 1987, 1991; Bacri and Salin, 1986). A calibration curve is first set up that plots the velocity variations as a function of the concentration of the mixture (of the solvent and solute). Theoretically, the velocity v_s of a fast compressional wave is given by

$$v_s = \sqrt{\frac{H}{\rho_e}}, \quad (11.21)$$

where

$$H = K_b + \frac{4}{3}\mu_s + \frac{(K_s - K_b)^2 K_f}{(1 - \phi) K_s K_f + \phi K_s^2 - K_b K_f}, \quad (11.22)$$

where K_b and μ_s are the bulk and shear moduli of the dry solid matrix of the porous medium, K_s and K_f are the bulk moduli of the porous medium and the fluid, and ϕ is the porosity. The quantity ρ_e is the effective density of the porous medium + the fluid, given by

$$\rho_e = \phi \rho_f + (1 - \phi) \rho_s, \quad (11.23)$$

with ρ_f and ρ_s being the densities of the fluid and solid, respectively. For the case of an unconsolidated porous medium, one has $K_b = \mu_s = 0$.

The sample is saturated by the solvent, the solute is then injected into the porous medium, and the sound wave velocity is measured at different cross sections (i.e., various axial positions x). The measurements are then transformed into equivalent solute concentrations $C(x, t)$. The typical accuracy for the $C(x, t)$ measurements are better than 1%. The results are then fitted to Eq. (11.12), with the fitting parameter being D_L .

11.5.2

Transverse Dispersion Coefficient

Measuring D_T is more difficult. If the porous medium is arranged as in Figure 11.3, then a mixed zone develops in the transverse direction. The concentration profile along a line perpendicular to the direction of the flow (the longitudinal direction) is typically S-shaped (see Figure 11.3). Then, D_T is determined by plotting percent composition versus the distance from the 50% composition on an arithmetic probability paper, and is estimated by the following equation,

$$D_T = \frac{v}{L} \left(\frac{z_{0.9} - z_{0.1}}{3.625} \right)^2, \quad (11.24)$$

where $z_{0.9}$ is the transverse distance between the 90 and 50% compositions. The reader should be able to derive Eq. (11.24).

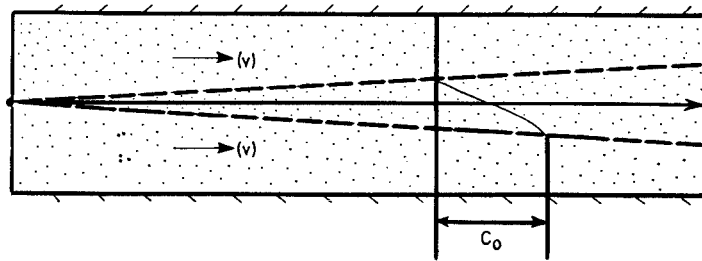


Figure 11.3 Schematic of a transverse dispersion experiment. The area between the dashed lines is the dispersion zone (after Harleman and Rumer, 1963).

11.5.3

Nuclear Magnetic Resonance Method

As described in Chapter 4, nuclear magnetic resonance (NMR) is a powerful tool for probing the structure of a porous medium. The same method can be used for measuring the dispersion coefficients as well as the effective diffusion coefficients in porous media that was already described in Chapter 10. One reason that NMR can be used for measuring the dispersion coefficients is that, as described above, such measurements typically involve the use of tracer particles of the solute and their motion in a flowing fluid in a porous medium. The NMR also provides a tracer method in that every single molecule is tagged noninvasively by its local precession frequency in a nonuniform magnetic field (Callaghan, 1991; Seymour and Callaghan, 1997).

Magnetic resonance imaging makes it possible to measure the distribution of the local velocity $\mathbf{v}(\mathbf{r}, t)$ (Guilfoyle *et al.*, 1992; Müller *et al.*, 1995; Kutsovsky *et al.*, 1996; Lebon *et al.*, 1996; Sederman *et al.*, 1998; Tessier and Packer, 1998). However, as pointed out by Khrapitchev and Callaghan (2003), because the spatial and temporal resolutions are constrained by the overall resolution of the instrument, one must make “trade offs” between the two. For example, if one settles for an *ensemble-averaged* signal for the sample as a whole (i.e., coarser spatial resolution), one gains optimal *temporal* resolution in return. The best way of achieving a balance between the two is through pulsed gradient spin echo-NMR (PGSE-NMR) that allows the analysis of ensemble-averaged mean-square displacements of the solute particles over a time interval. Recall that the dispersion coefficients are defined in terms of the first two moments of the probability distribution functions. In addition, since one typically uses a Lagrangian velocity (i.e., one in a moving coordinate system) for describing dispersion (for example, in Taylor–Aris dispersion theory a new coordinate, $x_1 = x - v_m t$ was used; see below), its use with a PGSE-NMR is natural because the method collects signals from moving tracers or particles.

We follow Khrapitchev and Callaghan (2003) and describe the PGSE-NMR method. Figure 11.4 shows how two variants of the PGSE-NMR are implemented in which the magnetic-field gradient $\mathbf{g}$ is given by $\mathbf{g} = \nabla M_z$, where M_z is the component of the magnetic field $\mathbf{M}$ parallel to $\mathbf{M}_0$, the homogeneous polarizing field, with $|\mathbf{M}_0| \gg |\mathbf{M}|$. As Figure 11.4 indicates, a sequence of short pulses of strength $G = |\mathbf{g}|$ are used in order to induce a phase shift (for every nuclear spin) that depends on the positions of its “mother” molecule in the gradient $\mathbf{g}$. The phase factors are $\exp(2i\pi \mathbf{q} \cdot \mathbf{r})$, with $\mathbf{r}$ being the position of the host molecule, and $\mathbf{q}$ the wavevector, $\mathbf{q} = (2\pi)^{-1} \gamma \mathbf{g} \delta$, where γ is the magnetogyric ratio and δ the duration of the gradient pulse G .

First, consider the single gradient pulse pair sequence, hereafter referred to as S-PGSE-NMR. If $\mathbf{r}(0)$ and $\mathbf{r}(\Delta)$ are, respectively, the positions of the spin-bearing molecule at the time of the first gradient pulse and the time at which a second pulse is applied a time Δ later, the accumulated spin phase is $\exp\{2i\pi \mathbf{q} \cdot [\mathbf{r}(\Delta) - \mathbf{r}(0)]\}$.

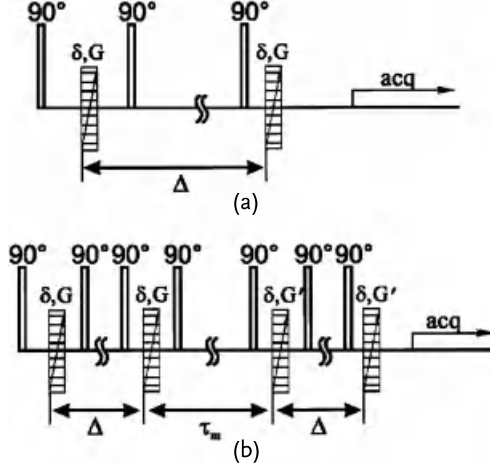


Figure 11.4 Implementation of the PGSE-NMR using stimulated echo rf pulse trains for (a) single PGSE-NMR, and (b) double PGSE-NMR (after Khrapitchev and Callaghan, 2003).

The normalized echo signal $E_S(q)$ is given by an ensemble average:

$$E_S(q) = \langle \exp\{2i\pi q \cdot [r(\Delta) - r(0)]\} \rangle. \quad (11.25)$$

Since $r(\Delta) - r(0) = \int_0^\Delta \mathbf{v}(t) dt$, we have

$$\begin{aligned} E_S(q) &= \left\langle \exp \left(2i\pi q \cdot \int_0^\Delta \mathbf{v}(t) dt \right) \right\rangle \\ &= \exp(2i\pi q \cdot \mathbf{v}\Delta) \left\langle \exp \left(2i\pi q \cdot \int_0^\Delta \mathbf{u}(t) dt \right) \right\rangle \\ &\approx 1 - 2\pi^2 q^2 \int_0^\Delta \int_0^\Delta \langle \mathbf{u}(t) \mathbf{u}(t') \rangle dt dt', \end{aligned} \quad (11.26)$$

where, $q = |q|$ and $\mathbf{v} = \mathbf{V} + \mathbf{u}$, with $\mathbf{V}$ being the ensemble-averaged velocity. Hence, $\mathbf{u}$ is the velocity fluctuations around $\mathbf{V}$. The dispersion coefficients D_* are then defined by

$$D_* = \frac{1}{4\pi^2 \Delta} \lim_{q \rightarrow 0} \frac{\partial}{\partial q^2} |\ln E_S(q)|, \quad (11.27)$$

where $*$ stands for L or T , and the D_* is measured for the velocity component of $\mathbf{u}(t)$ in the direction of $\mathbf{q}$. To measure the dispersion coefficients, the velocity $\mathbf{u}(t)$ is decomposed into longitudinal and transverse components. Thus, for example,

$$D_L = \frac{1}{2\Delta} \int_0^\Delta \int_0^\Delta \langle u_L(t) u_L(t') \rangle dt dt', \quad (11.28)$$

where $u_L(t)$ is the longitudinal component of $\mathbf{u}(t)$. A similar formula is used for D_T based on $u_T(t)$.

A similar analysis may be carried out for the second pulse sequence shown in Figure 11.4. Two gradient pairs are applied on the same spin magnetization, but over different time intervals, Δ , separated by a *mixing time* Δ_m . The displacement encoding for the two successive gradient pulse pairs can be applied in either the same or opposite sense. The experiments are labeled uncompensated and compensated, and we refer to the latter as Double PGSE-NMR or D-PGSE-NMR. Then,

$$E_D = \left\langle \exp \left(2i\pi \mathbf{q} \cdot \int_0^{\Delta} \mathbf{u}(t) dt \right) \exp \left(-2i\pi \mathbf{q} \cdot \int_{\Delta_m + \Delta}^{\Delta_m + 2\Delta} \mathbf{u}(t) dt \right) \right\rangle. \quad (11.29)$$

If the displacements during the two successive encoding intervals are completely uncorrelated, then the D-PGSE-NMR will yield twice the stochastic part of the exponent for the S-PGSE-NMR and, thus, $E_D = |E_S|^2$. Then, for the D-PGSE-NMR,

$$D_* = \frac{1}{4\Delta} \left[\int_0^{\Delta} \int_0^{\Delta} \langle \mathbf{u}(t) \mathbf{u}(t') \rangle dt dt' - 2 \int_0^{\Delta} \int_{\Delta_m + \Delta}^{\Delta_m + 2\Delta} \langle \mathbf{u}(t) \mathbf{u}(t') \rangle dt dt' + \int_{\Delta_m + \Delta}^{\Delta_m + 2\Delta_m} \int_{\Delta + \Delta_m}^{\Delta + 2\Delta} \langle \mathbf{u}(t) \mathbf{u}(t') \rangle dt dt' \right]. \quad (11.30)$$

Clearly, one decomposes $\mathbf{u}(t)$ into the longitudinal and transverse components in order to estimate D_L and D_T , in a manner similar to Eq. (11.28). If $\mathbf{u}(t)$ is stationary, then Eq. (11.30) is simplified to

$$D_* = \frac{1}{2\Delta} \left[\int_0^{\Delta} \int_0^{\Delta} \langle \mathbf{u}(t) \mathbf{u}(t') \rangle dt dt' - \int_0^{\Delta} \int_{\Delta_m + \Delta}^{\Delta_m + 2\Delta_m} \langle \mathbf{u}(t) \mathbf{u}(t') \rangle dt dt' \right]. \quad (11.31)$$

Note, however, that one must have $\langle \mathbf{u}(t) \rangle = 0$. Thus, one can write $\mathbf{u}(t) = \delta \mathbf{u} + \mathbf{u}_f(t)$, where $\mathbf{u}_f(t)$ is the fluctuating part of $\mathbf{u}$. Physically, $\delta \mathbf{u}$ can be interpreted as that part of $\mathbf{u}$ that is associated with *reversible* dispersion. In Taylor–Aris dispersion in a tube (see below), for example, if molecular diffusion is not significant, dispersion will be reversible, that is, by reversing the direction of the flow in the tube, the solute particles will reverse their paths exactly and return to their origin. In a D-PGSE-NMR experiment, the phase shift due to $\delta \mathbf{u}$ will be reversed in the second encoding and, thus, there will be no contribution. Thus, the dispersion coefficients will still be given by Eq. (11.30), except that $\mathbf{u}(t)$ should be replaced by $\mathbf{u}_f(t)$. That is not, of course, the case for S-PGSE-NMR experiments because in this case, the effect of $\delta \mathbf{u}$ is not eliminated. In fact, in this case,

$$D_*^{(S)} = \frac{1}{2} \langle \delta \mathbf{u}^2 \rangle \Delta + \frac{1}{2\Delta} \int_0^{\Delta} \int_0^{\Delta} \langle \mathbf{u}_f(t) \mathbf{u}_f(t') \rangle dt dt', \quad (11.32)$$

where the superscript S indicates a S-PGSE-NMR measurement.

To use the above formulation for estimating the dispersion coefficients, one must have detailed data on $\mathbf{u}(t)$. As mentioned earlier, PGSE-NMR experiments do provide such data. Then, the data are either used directly in the above information, or one fits a good model to the data, and utilizes the results in the formulation. An accurate model for such data is the Ornstein–Uhlenbeck process, which is stationary and given by

$$\langle \mathbf{u}(t) \mathbf{u}(0) \rangle = \langle \mathbf{u}^2 \rangle \exp\left(-\frac{t}{\Delta_c}\right), \quad (11.33)$$

where Δ_c is the correlation time. In principle, Δ_c is a distributed quantity. Moreover, the distribution of Δ_c for the longitudinal correlation times is not necessarily the same as that of the transverse ones. In addition, one must also consider two distinct regimes, namely, $\Delta \ll \Delta_c$ and $\Delta \gg \Delta_c$. After substituting Eq. (11.33) into Eqs. (11.30) and (11.32), and carrying out the integration, one obtains the desired expressions for the two dispersion coefficients. For $\Delta \ll \Delta_c$, one has

$$D_*^{(S)} \approx \frac{1}{2} \Delta (\langle \delta \mathbf{u}^2 \rangle + \langle \mathbf{u}_f^2 \rangle), \quad (11.34)$$

$$D_*^{(D)} \approx \frac{1}{2} \langle \mathbf{u}_f^2 \rangle \Delta \left[1 - \exp\left(-\frac{\Delta_m}{\Delta_c}\right) \right], \quad (11.35)$$

whereas for $\Delta \gg \Delta_c$, we obtain

$$D_*^{(S)} \approx \frac{1}{2} \langle \delta \mathbf{u}^2 \rangle + \langle \mathbf{u}_f^2 \rangle \Delta_c, \quad (11.36)$$

$$D_*^{(D)} \approx \langle \mathbf{u}_f^2 \rangle \Delta_c. \quad (11.37)$$

Seymour and Callaghan (1997), Stapf and Packer (1998), Khrapitchev and Callaghan (2003), and Hunter and Callaghan (2007) used the above and similar ideas to measure the dispersion coefficients by the PGSE-NMR method. In general, the measured dispersion coefficients are consistent with those measured by other methods (see Figures 11.5 and 11.6), but typically somewhat lower. Damion *et al.* (2000) carried out pore network simulations of dispersion (see below), and computed the important quantities in a NMR experiments using the model.

11.6

Dispersion in Systems with Simple Geometry

Dispersion is a general mass transfer phenomenon and can occur in a wide variety of systems. At the same time, dispersion is sensitive to the microstructure of the system in which it is occurring. Thus, dispersion has been studied in a large number of systems with a wide variety of morphologies. The simplest system in which dispersion can occur is in laminar flow through a single capillary tube, or in a channel between two parallel flat plates. The problem has been studied extensively, and in what follows, we summarize the main results. As we describe later

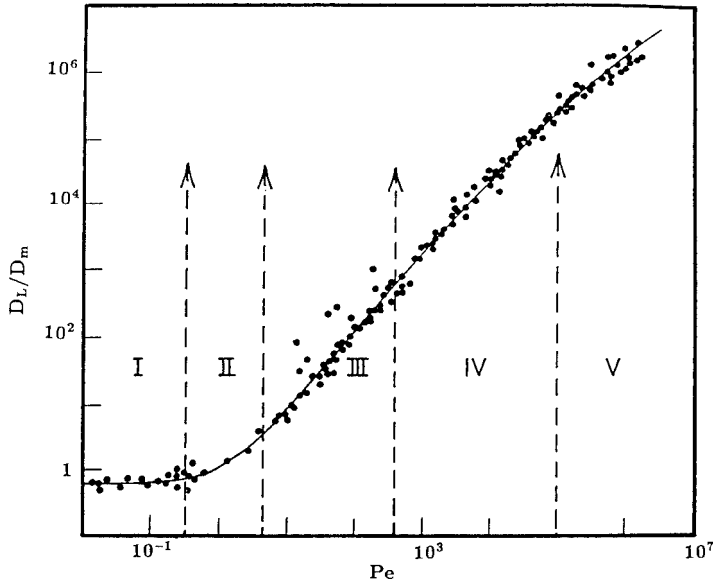


Figure 11.5 Dependence of the longitudinal dispersion coefficient D_L on the Péclet number Pe , and the various dispersion regimes. $D_m = D$ is the molecular diffusivity (after Fried and Combarrous, 1971).

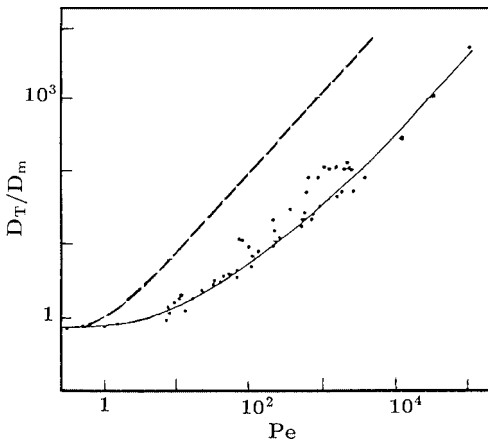


Figure 11.6 Dependence of the transverse dispersion coefficient D_T on the Péclet number Pe . For comparison, the $D_L - Pe$ curve (dashed) is also sketched (after Fried and Combarrous, 1971).

in this chapter, some of the results for dispersion in a single capillary tube are surprisingly similar and applicable to those in porous media with complex morphologies.

11.6.1

Dispersion in a Capillary Tube: The Taylor–Aris Theory

Historically, Griffiths (1911) was the first to report some experimental data that demonstrated the essence of the dispersion process in a tube with the effect of molecular diffusion being present. Griffiths did not, however, analyze the problem mathematically. He observed that a tracer solute injected into a tube in which water is flowing spreads out symmetrically about a plane in the cross section that moves with the speed of flow. He commented that, “*It is obvious that the movement of the center of the column of the tracers must measure the mean speed of flow.*” It turned out that this was not as obvious as Griffiths had thought!

Forty two years later, Taylor (1953)²⁾ pointed out that Griffiths’ observation is a rather startling result for two reasons. First, because the water at the center of the tube moves with twice the mean speed of the flow (the Hagen–Poiseuille flow), the water at (or near) the center must approach the column of the tracer, absorb the tracer as it passes through the column, and then reject it as it leaves on the other side of the column. Secondly, although the velocity is asymmetrical about the plane moving at the mean speed, the column of tracer spreads out *symmetrically*.

Taylor was known to have excellent intuition about the most important physical factors in any problem that he studied. He analyzed dispersion in a capillary tube approximately, but his analysis turned out later to be correct. Aris (1956)³⁾ studied the same problem without making any of the approximations that Taylor had made. They both studied dispersion in a cylindrical capillary tube of radius R . Starting from a CD equation for a tube

$$\frac{\partial C}{\partial t} + 2V_m \left[1 - \left(\frac{r}{R} \right)^2 \right] \frac{\partial C}{\partial x} = D_m \left(\frac{\partial^2 C}{\partial r^2} + \frac{1}{r} \frac{\partial C}{\partial r} + \frac{\partial^2 C}{\partial x^2} \right), \quad (11.38)$$

where V_m is the mean flow velocity in the tube, and defining a *mean* concentration C_m by

$$C_m = \frac{\int_0^{2\pi} \int_0^R C(r, x) r dr d\theta}{\int_0^{2\pi} \int_0^R r dr d\theta} = \frac{2}{R^2} \int_0^R C r dr, \quad (11.39)$$

- 2) *Sir Geoffrey Ingram Taylor* (1886–1975) was a British physicist and mathematician who made seminal contributions to the theory of turbulence and fluid dynamics, wave theory, solid mechanics, and dispersion among other things. He has been described as “one of the greatest physical scientists of the 20th century”.
- 3) *Rutherford “Gus” Aris* (1929–2005) was the Regents Professor of Chemical Engineering and Materials Science at the University of Minnesota. He finished a mathematics

degree from the University of London at the age of 16, and received a doctorate by correspondence, writing his thesis in six weeks. A member of the National Academy of Engineering, he made seminal contributions to the field of control theory as well as reaction engineering and catalysis. One of his best known quotes, the basis of dynamic programming is, “If you don’t do the best you can with what you happen to have, you’ll never do the best you might have done with what you should have had.”

Taylor and Aris showed that in the limit of long times,

$$\frac{\partial C_m}{\partial t} = D_L \frac{\partial^2 C_m}{\partial x_1^2}, \quad (11.40)$$

where $x_1 = x - V_m t$ is the moving coordinate with respect to the mean-flow velocity, and

$$D_L = D_a + \frac{R^2 V_m^2}{48 D_r}, \quad (11.41)$$

with the subscripts a and r signifying the fact that D_a and D_r are the contributions of the axial and radial molecular diffusion, respectively. That is, if we delete $\partial^2 C / \partial x^2$ from Eq. (11.38) (i.e., neglect axial diffusion), D_a will also be deleted from Eq. (11.41). Numerically, of course, $D_a = D_r = D_m$. Note that in the Taylor–Aris dispersion theory, D_L depends *quadratically* on V_m .

Equation (11.40) has a deep physical meaning: The average solute concentration spread out by a dispersion process in the solvent – by a combination of radial diffusion and axial convection – follows a diffusion equation and, hence, a Gaussian distribution, except that the effective diffusivity is *not* the molecular diffusivity D_m , but the dispersion coefficient that may be viewed as an effective diffusivity given by Eq. (11.41).

We define the Péclet number⁴ Pe by $Pe = R V_m / D_m = t_r / t_c$, where $t_r = R^2 / D_m$ is a time scale for significant radial diffusion, and $t_c = R / V_m$ is the convection time scale. Thus, Pe is simply a measure of the competition between radial diffusion and convection. Then, Eq. (11.41) is rewritten as

$$\frac{D_L}{D_m} = 1 + \frac{1}{48} Pe^2. \quad (11.42)$$

Aris (1956) also showed that for a tube with a cross section of *any* shape, one has

$$D_L = D_m + \delta_s \frac{l_s^2 V_m^2}{D_m}, \quad (11.43)$$

where l_s is a characteristic length scale of the tube, and δ_s is a numerical factor that depends on the shape of the tube's cross section. For example, for an elliptical cross section with the major and minor semi-axes a and b , one has $l_s = a$ and

$$\delta_s = \frac{1}{48} \frac{24 - 24e^2 + 5e^4}{24 - 12e^2}, \quad (11.44)$$

with $e = \sqrt{1 - b^2/a^2}$. For a circular cross section, $b = a$, $e = 0$ and $\delta = 1/48$, as expected. For dispersion in a slit pore with fully-developed laminar flow, $l_s = h$ and $\delta_s = 2/105$, where h is the half-width of the pore. Thus, the quadratic dependence of D_L on V_m is independent of the shape of the cross section. It is, in fact, the result of the competition between radial molecular diffusion and axial convection.

4) Jean Claude Eugene Péclet (1793–1857) published several books, including one on heat conduction.

Aris (1956) conjectured that *any* initial distribution of the solute concentration will ultimately approach a Gaussian distribution. Chatwin, in a series of papers (see Chatwin, 1977, for earlier references to his work), proved Aris' conjecture. The range of validity of the Taylor–Aris dispersion has been studied thoroughly by finite-difference calculations (Annanthakrishnan *et al.*, 1965) and by orthogonal collocation (Wang and Stewart, 1983).

Further important work on dispersion in tubes was carried out by Horn (1971) and Brenner (1980). In particular, Brenner (1980) generalized the Taylor–Aris dispersion theory very significantly by employing a formulation in which both local and global spaces are utilized. For example, in the problem of dispersion in a capillary tube, r represents the local space, whereas x is the global one. Brenner also assumed that the solute particles may have a finite size. One can also exploit the equivalence between Langevin and Fokker–Planck equations in order to derive the Taylor–Aris results (van den Broeck, 1982).

Experiments by Koutsky and Adler (1964) demonstrated that coiled tubes yield reduced longitudinal dispersion, which is important in chemical reactor design as well as in diffusivity measurements. Hoagland and Prud'homme (1985) analyzed longitudinal dispersion in tubes of sinusoidally-varying radius, $R(z) = R_0[1 + a \sin(2\pi z/\omega)]$, in order to model dispersion in packed-bed processes. Surprisingly, their analysis indicated that the longitudinal dispersion coefficient D_L varies *linearly* with the Péclet number, rather than the second power as in the classical Taylor–Aris dispersion.

11.6.2

Dispersion in Spatially-Periodic Models of Porous Media

At the next level of complexity are spatially-periodic models of porous media. We already described the geometrical structure of such models in Chapter 5. Diffusion, electrical conduction, and flow in such models of porous media were studied in Chapter 9. Thus, it suffices to mention that Brenner (1980), Brenner and Adler (1982), Eidsath *et al.* (1983), Koch *et al.* (1989), and Salles *et al.* (1993) examined theoretically dispersion in spatially-periodic 3D porous media, while Edwards *et al.* (1991) studied it within the 2D model. Edwards (1995) studied charge transport together with convective dispersion in spatially-periodic models. Dorfman and Brenner (2002) studied Taylor–Aris dispersion for Brownian particles of nonzero sizes in a spatially-periodic network of capillary tubes. Quintard and Whitaker (1993) developed a method for studying transport and dispersion in spatially-disordered porous media with or without disorder. Brenner and Edwards (1993) presented analyses of convective dispersion and reaction in various geometries, including tubes and spatially-periodic packed beds. On the experimental side, Gunn and Pryce (1969) reported measurements of the longitudinal dispersion coefficient D_L for flow parallel to one of the axes of a simple-cubic lattice of spherical particles.

Such models are not realistic representations of real porous media that are neither periodic, nor ordered the way such models are. In fact, because dispersion is very sensitive to the microstructure of the system in which it occurs, a spatially-

periodic model of porous media cannot be expected to yield useful predictions for disordered porous media. Indeed, Koch *et al.* (1989) showed, for example, that for a square array of cylinders or a cubic array of spheres and in the limit $Pe \rightarrow \infty$, D_L depends quadratically on Pe , and that D_T approaches a *constant value*. Their predictions are in contradiction with dispersion in *disordered* porous media for which D_L has a much weaker dependence on Pe , and D_T does not reach a constant value (see below).

11.7

Classification of Dispersion Regimes in Porous Media

Over the past several decades, dispersion in porous media has been studied experimentally, particularly in beadpacks, unconsolidated sandpacks, and sandstones. Some of the reported data, mainly for unconsolidated sands, were compiled by Fried and Combarous (1971). Data on sandstones, compiled by Perkins and Johnston (1963) and Legaski and Katz (1967) show, however, that dispersion in consolidated porous media is similar to that in unconsolidated media. Figure 11.5 collects experimental data for D_L/D_m for sandpacks, which show that there are five discernible regimes of dispersion; see Bijeljic *et al.* (2004) for a collection of the more recent references on the experimental data for D_L and its dependence on Pe .

Figure 11.6 presents the typical experimental data for D_T/D_m . For unconsolidated porous media, the Péclet number is defined as $Pe = d_g V/D_m$, where d_g is frequently taken to be the average diameter of a grain or bead. Bijeljic and Blunt (2007) provide a more recent list of references for the experimental measurements of D_T and its dependence on Pe .

The five dispersion regimes, indicated by Figure 11.5, are as follows.

1. $Pe < 0.3$ defines the *diffusion* regime in which convection is so slow that molecular diffusion controls the mixing almost completely. In this regime, we have *isotropic* dispersion such that (Brigham *et al.*, 1961)

$$\frac{D_L}{D_m} = \frac{D_T}{D_m} = \frac{1}{F\phi}, \quad (11.45)$$

where F is the formation factor and ϕ is the porosity of the porous medium. The quantity $1/(F\phi)$ varies commonly between 0.15 and 0.7, depending on the type of porous media. Due to the isotropy, a concentrated sphere of solute will remain a sphere (rather than developing into an ellipsoid as indicated by Eq. (11.3)), but will increase in size as dispersion progresses. Although Eq. (11.45) is quoted widely in the literature, a proof of it, even by a heuristic argument, is not usually given. In particular, the presence of ϕ is not obvious. However, if we consider the limit $Pe = 0$, then we derive Eq. (11.45) by the following argument.

As mentioned in Chapter 3, Einstein's relation relates the electrical conductivity g_f of a fluid to the molecular diffusivity D_m by $g_f = ne^2 D_m/(k_B T)$, where n

is the density of the charge carriers, e is the charge, k_B is the Boltzmann's constant, and T is the temperature. For a porous medium, the same equation may be used, except that g_f should be replaced by g_e , the effective electrical conductivity of the fluid-saturated medium, D_m by D_e , the effective diffusivity in the porous medium, and n by $n\phi$, the density of charge carriers in the pore space of the medium and, thus, $g_e = n\phi e^2 D_e / (k_B T)$. Thus, $D_e/D_m = g_e/(\phi g_f) = 1/(F\phi)$, with $F = g_f/g_e$, which is equivalent to Eq. (11.45), because in the limit $Pe \rightarrow 0$, we have $D_L = D_T = D_e$.

2. The regime $0.3 < Pe < 5$ defines the *transition* zone in which convection contributes to dispersion, but the effect of diffusion is still quite strong. D_L/D_m appears to increase with Pe , although it is difficult to say how.
3. The interval $5 < Pe < 300$ defines the *power-law* regime. Convection dominates dispersion, but the effect of molecular diffusion cannot be neglected, and one writes

$$\frac{D_L}{D_m} = \frac{1}{F\phi} + a_L Pe^{\beta_L}, \quad (11.46)$$

$$\frac{D_T}{D_m} = \frac{1}{F\phi} + a_T Pe^{\beta_T}. \quad (11.47)$$

The *average* values of β_L and β_T from all the available experimental data are $\beta_L \simeq 1.2$ and $\beta_T \simeq 0.9$. We call this regime the *boundary-layer* dispersion after Koch and Brady (1985) since, as we show below, it is consistent with the existence of a diffusive boundary layer near the solid surface of the pores, first identified by Saffman (1959), where molecular diffusion transfers the solute from the very slow regions near the solid walls to faster streamlines. In practical applications, β_L and β_T often are taken to be unity. The coefficients a_L and a_T depend on the heterogeneities of the pore space, and their typical values are $a_L \simeq 0.5$ and $a_T \simeq 0.01$ – 0.05 .

4. The *purely convective regime* is defined by $300 < Pe < 10^5$. Simple dimensional analysis indicates that (ignoring the $1/(F\phi)$ term, which is small)

$$\frac{D_L}{D_m} \sim Pe, \quad (11.48)$$

$$\frac{D_T}{D_m} \sim Pe. \quad (11.49)$$

This type of dispersion is also called *mechanical dispersion*. In this case, dispersion is simply the result of a stochastic fluid velocity field induced by the randomly distributed pore boundaries.

5. For $Pe > 10^5$, dispersion is in the *turbulent* regime. The Péclet number is no longer the only correlating parameter, as the Reynolds number should also be considered. For flow through porous media, however, this regime is not of interest.
6. There is a sixth dispersion regime that is not evident in Figure 11.5. This is the so-called *holdup dispersion* (Koch and Brady, 1985) first studied by Carberry

and Bretton (1958), Turner (1959), and Aris (1959). In this regime, the solute is trapped in the dead-end pores or inside the solid grains, from which it can escape only by molecular diffusion. One has (again, ignoring the $1/(F\phi)$ term)

$$\frac{D_L}{D_m} \sim \text{Pe}^2, \quad (11.50)$$

$$\frac{D_T}{D_m} \sim \text{Pe}^2, \quad (11.51)$$

which indicate rather strong dependence of D_L and D_T on Pe . In low-porosity porous media that are barely connected with their porosity being close to the percolation threshold, there are many dead-end pores and, therefore, this regime is relevant to dispersion in such porous media.

Bacri *et al.* (1987) used an acoustic technique (see Section 11.5.1.3) to measure D_L in flow through three different porous media, namely, an unconsolidated pack of glass beads, a fireproof brick, and a mill sandstone. Their data indicated that pore-level disorder strongly affects D_L and its dependence on Pe . They also reported that their data follow power laws (11.46)–(11.49), depending on the breadth of the pore size distribution and the connectivity of the pore space.

11.8

Continuum Models of Dispersion in Porous Media

Having gained a qualitative understanding of dispersion in porous media and what we may expect, let us now describe and discuss various models of dispersion in porous media. We begin by describing the continuum approach.

11.8.1

The Volume-Averaging Method

Chapter 9 presented the continuum approach for modeling of flow and transport in porous media. The same approach may be used for studying dispersion in porous media. The works of Whitaker (1967), Bachmat (1969), Gray (1975), Carbonell and Whitaker (1983), Eidsath *et al.* (1983), and Plumb and Whitaker (1988a) fall in this class of methods. For example, Plumb and Whitaker (1988a) used a volume-averaging technique (see Whitaker, 1999, for details of the method) in order to average the microscopic CD equation, where the averaging is over the disorder, in order to arrive at the macroscopic averaged equation. The final result for the volume-averaged concentration $\langle C \rangle$ of the solute is given by

$$\phi \frac{\partial \langle C \rangle}{\partial t} + \nabla \cdot (\phi \langle \mathbf{v} \rangle \langle C \rangle) = \nabla \cdot (\phi \mathbf{D}_* \cdot \nabla \langle C \rangle). \quad (11.52)$$

Here, $\mathbf{D}_*$ is the dispersion coefficient tensor given by

$$\mathbf{D}_* = D_m \left[\mathbf{I} + \frac{1}{\mathcal{V}} \int_A \mathbf{n} \cdot \mathbf{f} dA \right] - \langle \tilde{\mathbf{v}} \mathbf{f} \rangle, \quad (11.53)$$

where $\mathbf{f}$ is a trial solution, $\mathcal{V}$ is the pore volume, A is the interfacial area contained between the averaging volume, and $\mathbf{n}$ is the unit outwardly-directed normal vector for the pores. Here, $\tilde{\mathbf{v}}$ is the fluctuations in the velocity $\mathbf{v}$ (i.e., $\mathbf{v} = \langle \mathbf{v} \rangle + \tilde{\mathbf{v}}$).

The volume-averaging analysis indicates that on a large enough length scale over which the porous medium is homogeneous, a CD equation describes the *average* solute concentration. The tensor $\mathbf{D}_*$ contains two terms. One is the contribution of molecular diffusion, while the second one is due to hydrodynamic transport. However, the contribution of molecular diffusion appears only as D_m , not as $D_m(F\phi)^{-1}$, as the method of volume-averaging cannot take into account the effect of the tortuosity of a porous medium.

The unknown function $\mathbf{f}$ must be determined in order for $\mathbf{D}_*$ to be computed. In practice, $\mathbf{f}$ is the solution of the following boundary value problem

$$\tilde{\mathbf{v}} + \mathbf{v} \cdot \nabla \mathbf{f} = D_m \nabla^2 \mathbf{f}, \quad (11.54)$$

$$-\mathbf{n} \cdot \nabla \mathbf{f} = \mathbf{n} \text{ at } A, \quad (11.55)$$

provided that $D_m t / l^2 \gg 1$ (where l is the length scale associated with the pores) and, therefore, $\mathbf{f}$ is determined if a model of the pore space is specified. If, however, the pore space is disordered, then the numerical calculation of $\mathbf{f}$ is no easier than the numerical simulation of the CD equation itself within the domain of the problem.

11.8.2

The Ensemble-Averaging Method

Koch and Brady (1985) used an ensemble-averaging technique (see also Chapter 9) in order to study dispersion in a packed bed in which the volume fraction of the porous particles was ϕ_p . They first formally related the average concentration field to the probability distribution of the solid material, and then derived the effective dispersion coefficients in the *high porosity limit* (low solid volume fraction ϕ_p), including all the relevant proportionality constants. Koch and Brady (1985) showed that the macroscopic equation of mass conservation (i.e., the CD equation) in the long-time limit takes the form of a macroscopic Fick's (second) law with a constant effective diffusivity, or dispersion, tensor. They also carried out an asymptotic analysis in low volume fraction of the dispersion coefficients in a bed of fixed spherical particles for all values of the Péclet number. Koch and Brady (1985) identified several physical mechanisms that cause dispersion:

1. The stochastic velocity fluctuations induced in the fluid by the randomly positioned bed particles give rise to a convectively-driven contribution to disper-

- sion. At high Péclet numbers, the convective dispersion mechanism is purely mechanical, with the resulting dispersion coefficients being independent of molecular diffusion and grow linearly with Pe .
2. The region of zero velocity in and near the particles gives rise to nonmechanical dispersion mechanisms that dominate the longitudinal dispersion coefficient at very high Péclet numbers. One such mechanism involves the retention of the diffusing solute particles in permeable particles, from which they can escape only by molecular diffusion, leading to a dispersion coefficient that grows as Pe^2 (see the earlier discussion of holdup dispersion)
 3. Even if the particles are impermeable, nonmechanical contributions that grow as $Pe \ln Pe$ and Pe^2 at high Pe arise from a diffusive boundary layer near the pores' surfaces and from regions of closed streamlines (the dead-end pores), respectively.

These results are all in agreement with the experimental observations described in the last section. As an example, the predictions of Koch and Brady (1985) for relatively high Péclet numbers are given by

$$\frac{D_L}{D_m} = 1 + \frac{3}{4}Pe + \frac{\pi^2}{6}\phi_p Pe \ln Pe + \frac{D_m(1+\chi)^2}{15\kappa D_{mp}}\phi_p Pe^2, \quad (11.56)$$

$$\frac{D_T}{D_m} = 1 + \frac{63\sqrt{2}}{320}\sqrt{\phi_p}Pe. \quad (11.57)$$

Here, κ is the partition coefficient for the solute (i.e., partitioning of the solute between the fluid flowing through the bed and that in the void space inside the particles), and D_{mp} is the molecular diffusivity of the solute inside the particles. χ is defined by

$$\chi = \frac{\phi_p(\kappa - 1)}{\kappa - \phi_p(\kappa - 1)}. \quad (11.58)$$

An appealing aspect of Koch and Brady's work was that the fluid mechanical aspects of the problem were treated *without* any approximations. The ensemble-averaging technique may also be extended to the case where dispersion is not Gaussian and cannot be described by a CD equation (see below).

11.9

Fluid-Mechanical Models

The fluid-mechanical models are based on three basic attributes:

1. a Lagrangian description of the motion of solute-containing fluid through a single pore. As mentioned earlier, in a Lagrangian approach, the motion of solute particles is followed, and the average velocity and the dispersion coefficients are defined as the time-rate of change of the mean and mean-square positions of the particles, respectively;

2. specific assumptions about the medium, for example, homogeneity and isotropy, and
3. calculation of the quantities of interest as statistical averages.

The works of Scheidegger (1954), Day (1956), de Josselin de Jong (1958), Saffman (1959)⁵, Saffman (1960), Haring and Greenkorn (1970), and Bear (1972) are in this group. Saffman's work is the most general of these and, hence, is described here. His model consisted of a network of randomly oriented and distributed straight capillaries, in each one of which the flow was uniform. The path of solute particles was regarded as a random walk (see Chapters 9 and 10) in which the length, direction, and duration of each step were random variables. However, the only morphological disorder allowed in the model was the random orientations of the capillary tubes. Saffman was careful to introduce a dynamical basis for his model founded explicitly on fluid-mechanical aspects. He assumed that all the pores have an equal circular cross section of radius R , that flow was laminar in all the tubes, and distinguished five cases in his first paper Saffman (1959):

1. $t_c \ll t_r$, where t_c is the convective time spent by a solute particle in a pore and t_r the time required for appreciable radial diffusion of the same particle, $t_r = R^2/(8D_m)$ (i.e., the time that the solute particle spends to jump a distance $R/2$ from one streamline to another). Thus, radial diffusion is negligible and the duration of a step is $t = t_c = l/V_m$, where l is the pores' length.
2. The solute particle is on a streamline close to a pore wall, that is, its speed is small and molecular diffusion is important. The duration of a step is then $t = t_r + l/V_m$, that is, the particle makes *one jump* from a streamline close to the pore wall to another one with speed V_m , and then is convected out of the pore.
3. $t_r < t \ll t_a$, where t_a is the time for appreciable *axial* diffusion with $t_a = l^2/(2D_m)$. The effect of the axial diffusion is negligible and $t = t_r + l/V_m$.
4. $t_r < t \leq t_a$, which means that the pore is very narrow and thus, $t = l/V_m$.
5. $t_a \ll t$. Therefore, the duration of a step is $t = t_a$.

Saffman found that in all the cases, D_T is given by

$$D_T = \frac{3}{16} l V . \quad (11.59)$$

D_L was found, however, to depend on the dispersion regime considered. If we let

$$D_L = \frac{1}{2} V l s^2 , \quad (11.60)$$

- 5) *Philip Geoffrey Saffman* (1931–2008), the Theodore von Karman Professor of Applied Mathematics and Aeronautics at California Institute of Technology, was a leading figure in fluid mechanics. In addition to his work on dispersion and the Saffman–Taylor

instability (see Chapter 13), he developed, together with his colleague Max Delbrück, the Saffman–Delbrück model of protein diffusion, and made significant contributions to the theory of fluid vorticity arising from the motion of ships and aircrafts.

then

$$s^2 = \frac{1}{3} \ln \left(\frac{3Vt_a}{l} \right) + \frac{1}{12} \left[\ln \left(\frac{6Vt_r}{l} \right) \right]^2 - \frac{1}{4} \ln \left(\frac{6Vt_r d}{l} \right) + \frac{19}{24}, \quad (11.61)$$

if

$$\frac{\frac{Vt_a}{l}}{\sqrt{n_s \ln \left(\frac{3Vt_a}{l} \right)}} \ll 1; \quad (11.62)$$

and

$$s^2 = \frac{1}{6} \ln \left(\frac{27Vt_m}{2l} \right) + \frac{1}{12} \left[\ln \left(\frac{6Vt_r}{l} \right) \right]^2 - \frac{1}{4} \ln \left(\frac{6Vt_r}{l} \right) + \frac{19}{24}, \quad (11.63)$$

if

$$\ln n_s \gg 2, \quad \frac{\frac{3Vt_r}{l}}{n_s^{\frac{1}{2}}} \ln n_s^{\frac{1}{2}} \ll 1, \quad \frac{\frac{3Vt_a}{l}}{n_s^{\frac{1}{2}}} \ln n_s^{\frac{1}{2}} \gg 1,$$

and

$$s^2 = \frac{1}{48} \left[\ln \left(\frac{54Vt_m}{l} \right) \right]^2, \quad (11.64)$$

if

$$\ln n_s \gg 2, \quad \frac{\frac{4Vt_r}{l}}{n_s^{\frac{1}{2}}} \ln n_s^{\frac{1}{2}} \gg 1, \quad \frac{\frac{4Vt_a}{l}}{n_s^{\frac{1}{2}}} \ln n_s^{\frac{1}{2}} \gg 1,$$

where n_s is the mean number of steps taken by the fluid particles after a large time T_m at which D_L and D_T are measured, given by $T_m = 3\langle x \rangle / (2l)$ with $\langle x \rangle$ being the mean longitudinal position of the particles at time T_m . We may view the constant 19/24 in Eqs. (11.61) and (11.63) as the prediction of the Saffman's theory for $(F\phi)^{-1}$. Ignoring the constant that is usually much smaller than the other terms in the above equations, Saffman's results for D_L may be summarized as

$$\frac{D_L}{D_m} \sim \text{Pe}(\ln \text{Pe})^\alpha, \quad (11.65)$$

where $\alpha = 1$ or 2. Equation (11.65) may now be compared with Eq. (11.46). As first pointed out by Sahimi (1984), if we take $\alpha = 1$ and fit the experimental data to Eq. (11.65), the resulting fit would be as accurate as that provided by Eq. (11.46) if $\beta_L \simeq 1.25$. On the other hand, if we take $\alpha = 2$, the accuracy of the resulting fit would be compatible with that of Eq. (11.46) if $\beta_L \simeq 1.15$. Thus, two important features of all the experimental data are explained:

1. The data indicate that β_L is either about 1.13–1.16 (obtained by Legaski and Katz (1967) for Bandera sandstone, by Salter and Mohanty (1982) for Berea sandstone, and by Blackwell *et al.* (1959) for unconsolidated packed sands), or about 1.24–1.30 (as reported by Brigham *et al.* (1961) and Pakula and Greenkorn (1971) for glass beads, and by Legaski and Katz (1967) for Boise and Nordosaria sandstones and for Dolomites), with an overall average of about 1.2, as mentioned above.
2. β_L is probably not universal; it depends on the strength of the competition between molecular diffusion and convection, which in turn depends on the pore shapes and sizes. On the other hand, Eq. (11.59) is *not* completely compatible with Eq. (11.47) because most of the data (see, for example, Blackwell, 1962) indicate that $\beta_T \simeq 0.9$, as mentioned above. Although, in the numerical simulations, β_T is usually taken to be unity. Similar to β_L , it is likely that β_T is *not* universal.

Saffman also found that dispersion *cannot* be described by a CD equation unless the characteristic time T_m is sufficiently large. His analysis clearly points to the significance of molecular diffusion to dispersion in microscopically-disordered porous media, no matter how small it may be, as long as it is not exactly zero. In fact, the presence of molecular diffusion in such porous media is essential. In its absence, a solute particle that is traveling along a streamline very close to a pore wall will need a huge amount of time to escape from that region. Otherwise, $D_L = 0$. It is molecular diffusion that intervenes and transfers the solute to a faster streamline and prevents D_L from vanishing. The logarithmic terms in Eqs. (11.61)–(11.64) are precisely due to the diffusion time scale for transferring the solute from a very slow streamline to a faster one.

Saffman's results are presumably valid if Pe is large but finite. In his second paper (Saffman, 1960), he considered the case where Pe is "less than some large value", and found that both D_L and D_T depend quadratically on Pe . The agreement between Saffman's results and the various experimental data ranges from reasonable to good. Saffman's work is the most detailed and carefully analytical analysis of dispersion in microscopically disordered porous media. It has not, however, been fully appreciated.

Saffman did not allow the possibility that the pores' sizes are distributed according to a pore size distribution (all the pores were assumed to have the same radius). Haring and Greenkorn (1970) rederived some of Saffman's results, assuming that the pores' sizes follow a statistical distribution. Moreover, in Saffman's work, the flow field is represented by a sort of a mean-field approximation. There are no correlations between the successive steps of the particle. This restriction can also be removed by Monte Carlo calculations using pore network models of pore space, as was first done by Sahimi *et al.* (1982), which is described below. Finally, it is worth mentioning that the logarithmic singularities derived by Saffman were re-discovered by Aronovitz and Nelson (1984) in what they called "diffusion in steady flow" through a porous medium, which is nothing but hydrodynamic dispersion described herein.

11.10

Pore Network Models

These models belong to the class of fluid-mechanical models that we already described, except that the mean-field nature of the flow field and the absence of the heterogeneity are explicitly deleted. As already mentioned in Chapter 3, Torelli and Scheidegger (1972) appear to be the first to propose a random network model for studying dispersion processes in porous media (and invoked percolation theory to explain the phenomenon), although they did not report any result. Torelli (1972) did simulate dispersion processes in flow through a random network, but his results pertain to a type of non-Gaussian transport process not closely related to what we are interested in here.

11.10.1

First-Passage Time and Random Walk Simulation

Sahimi *et al.* (1982) were the first to use pore network models of porous media to simulate dispersion in flow through the networks. In their model, one first determines the flow field in the network by the method described in Chapter 10, where we described pore network models for calculating the effective permeability of a porous medium. Then, solute particles are injected into the network at random at the upstream plane at $x = 0$. Each particle selects a streamline at random. The convective travel time for a given pore is given by $t = l/v_p$, where l is the pore's length, and v_p is the flow velocity in the pore. Complete mixing at the nodes was assumed and, therefore, the probability that a pore is selected once a particle has arrived at a node is proportional to the flow rate in that pore. The FPTD Q for the particles are computed by fixing the longitudinal or lateral positions and measuring the time at which the particles arrive at the fixed positions for the first time. It is not difficult to show that D_ζ , the dispersion coefficient in the ζ -direction, is given by

$$D_\zeta = \int_0^\infty Q(\zeta - \zeta_0, t) \frac{s_\zeta^2}{2t} dt, \quad (11.66)$$

where $\zeta_0(x_0)$ is the starting position of the particles, $s_x^2 = (x - x_0 - Vt)^2$, and, $s_\zeta^2 = (\zeta - \zeta_0)^2$ for $\zeta = y$ and z .

However, such a model is appropriate for purely mechanical dispersion since it ignores the pore-level molecular diffusion. It only considers the effect of a stochastic velocity field throughout the network. To include the effect of molecular diffusion and simulate the boundary-layer dispersion, the following method was adopted (Sahimi and Imdakm, 1988). The convective time t_c for traveling along a streamline in a pore is calculated. If $t_c \gg t_r$, where t_r is the time scale for significant radial diffusion that was described above, then one sets $t = t_c + t_r$ since the tracer has enough time to diffuse to a faster streamline. Sorbie and Clifford (1991) suggested a somewhat different method for including the effect of molecular diffusion

on the dispersion process in the pore network. To simulate the holdup dispersion (i.e., dispersion in which the tracer may diffuse into the stagnant region), the tracer particles are allowed to diffuse into the dead-end pores of the network. Transport in such pores is only by molecular diffusion. In a series of papers, Sahimi *et al.* (1982, 1983a, 1986a,b) and Sahimi and Imdakm (1988) demonstrated that such pore network models reproduce and simulate all the regimes of dispersion described above. In particular, Eqs. (11.33)–(11.38) are all correctly predicted by such FPT simulations and pore network models.

A more refined pore network model of dispersion using random walk simulations was developed by Bijeljic *et al.* (2004). They mapped a Berea sandstone onto an equivalent pore network with square cross sections, and utilized the random walk simulation similar to that developed by Sahimi and co-workers. To include the effect of molecular diffusion, the particles are moved randomly, as defined by the coordinates of a spherical coordinate system. In a time step Δt , the diffusion distance is $r_d = \sqrt{2D_m \Delta t}$. Thus, the coordinates of the particle are given by the usual relation, $x = r_d \cos \theta \sin \varphi$, $y = r_d \sin \theta \sin \varphi$, and $z = r_d \cos \varphi$. The transition probabilities for moving the particles from one pore to another depended on the last step of the particles. If they arrived at a node of a network as a result of convection, then the next pore was, similar to the work of Sahimi and co-workers, selected with a probability proportional to the volume flow rate in that pore. If the particles arrived at a node as a result of a diffusion step, then the probability of selecting the next pore was taken to be proportional to the cross-sectional area of that pore. The longitudinal dispersion coefficient D_L was then computed by the method that Sahimi and co-workers used. Bijeljic *et al.*'s simulations indicated that their network model correctly reproduces the dependence of D_L on the Péclet number Pe , as shown in Figure 11.5. A somewhat similar model was also used by Bruderer and Bernabé (2001) in their studies of the transition from Taylor–Aris dispersion to purely mechanical dispersion in very heterogeneous porous media; see also Acharya *et al.* (2007).

A similar model was used (Bijeljic and Blunt, 2007) for computing the transverse dispersion coefficient D_T . As in the case of D_L , the computed dependence of D_T on the Péclet number Pe was found to be similar to what is presented in Figure 11.6.

11.10.2

Probability Propagation Algorithm

De Arcangelis *et al.* (1986) proposed another pore network model that they called the *probability propagation algorithm*. In their model, a 1D CD equation is assumed to describe mass transfer in each pore of the network, that is,

$$\frac{\partial C}{\partial t} + V_m \frac{\partial C}{\partial x} = D_m \frac{\partial^2 C}{\partial x^2}. \quad (11.67)$$

Consider a network of capillary tubes $\{i, j\}$. The concentration C_{ij} in each tube follows Eq. (11.67), with the initial condition that $C_{ij}(x_{ij}, 0) = 0$, and three boundary conditions:

1. a unit pulse of input flux at node i at time $t = 0$,

$$\sum_{\{j\}} S_{ij} \left(\mathbf{v}_{mij} C_{ij} - D_m \frac{\partial C_{ij}}{\partial x_{ij}} \right)_{x_{ij}=0} = \delta(t), \quad (11.68)$$

where S_{ij} is the cross-sectional area of tube ij , and $\mathbf{v}_{mij}$ is the mean-flow velocity in that tube;

2. a common concentration $C_i(t)$ at the starting junction, $C_{ij}(0, t) = C_i(t)$ for all j , and
3. a sink at each tube end, $C_{ij}(l, t) = 0$, for all j , corresponding to the fact that a tracer reaching the end acts as a source for the junction problem at the new node.

The FPT probability is given by $q_{ij}(t) = -S_{ij} D_m \partial C_{ij}(l, t) / \partial x_{ij}$. Equation (11.67) is solved in the Laplace transform space. The solution is given by

$$\tilde{C}_{ij}(x, \lambda) = A_{ij} \exp(\alpha_{ij} x) + B_{ij} \exp(\beta_{ij} x), \quad (11.69)$$

with

$$\alpha_{ij}, \beta_{ij} = \frac{v_{ij} \pm \sqrt{v_{ij}^2 + 4D_m \lambda}}{2D_m}, \quad (11.70)$$

where A_{ij} and B_{ij} are determined from the specified boundary conditions, and λ is the Laplace transform variable conjugate to t . We then obtain

$$\tilde{q}_{ij}(\lambda) = \tilde{C}_{ij}(\lambda) S_{ij} \frac{\alpha_{ij} - \beta_{ij}}{\exp(-\beta_{ij} l) - \exp(-\alpha_{ij} l)}. \quad (11.71)$$

Having determined $\tilde{q}_{ij}(\lambda)$, one obtains the FPTD $\tilde{Q}(L, \lambda)$ for the *entire* network,

$$\tilde{Q}(L, \lambda) = \sum_{\Gamma} \prod_{i,j \in \Gamma} \tilde{q}_{ij}(\lambda), \quad (11.72)$$

where the sum is over *all* the paths Γ from the inlet to the outlet of the network.

To efficiently compute the sum in Eq. (11.72), de Arcangelis *et al.* (1986) ordered the nodes of the network in decreasing pressure, starting with the inlet and finishing with the outlet. At each node i , a quantity $\tilde{Q}_i(\lambda)$ was introduced that represented a partial sum in Eq. (11.72), over paths running from the inlet to site i . For a delta-function input of the tracer, one initially has $\tilde{Q}_I = 1$ at the inlet I and $\tilde{Q}_i = 0$ elsewhere. One then proceeds recursively through the pressure-ordered node list, propagating the quantity $\tilde{Q}_i$ from each node i to its network neighbors j according to the rule that $\tilde{Q}_j(\lambda) \rightarrow \tilde{Q}_j(\lambda) + \tilde{Q}_i(\lambda) \tilde{q}_{ij}(\lambda)$, $\tilde{Q}_i(\lambda) \rightarrow 0$. After all the internal nodes have been propagated once in this way, the quantity $\tilde{Q}_0(\lambda)$ at the outlet contains all terms of the sum in Eq. (11.72) corresponding to purely downstream paths.

However, because molecular diffusion is present, the solute motion includes upstream paths as well. Hence, after one sweep through the network, one has $\tilde{Q}_n \neq 0$ for the internal nodes n . By repeated sweeps through the network, the contributions of the paths with progressively more upstream steps are included. Once $\tilde{Q}(L, \lambda)$ is determined, it is inverted to the time domain and D_L is calculated using Eq. (11.66). Note that this model, in the mechanical dispersion regime (i.e., with no boundary-layer diffusion), is equivalent to the FPT model of Sahimi and co-workers. De Arcangelis *et al.* (1986) showed that their method reproduces the results for both mechanical and boundary-layer dispersion. The method is very efficient as long as the network is well connected. For percolation networks near the percolation threshold, the method is very inefficient because calculating the sum in Eq. (11.72) becomes very time consuming.

11.10.3

Deterministic Models

Koplik *et al.* (1988b) developed another method for studying dispersion in pore networks. In their model, one first calculates the flow field throughout the pore network by the method described in Chapter 10. Assuming that dispersion in each pore is described by a CD equation, Eq. (11.68) with its right-hand side being zero (which is simply a statement of the continuity of mass at each node), is written for all the interior nodes of the pore network. The resulting set of linear equations for nodal concentrations is solved (in the Laplace transform space), from which D_L is calculated. Sahimi and Jue (1989) and Sahimi (1992a) used a similar method to study generalized Taylor–Aris dispersion of *finite-size* molecules in porous media, that is, one in which the hydrodynamic radius of the molecules is comparable to the pore sizes.

Roux *et al.* (1986) used the same method, except that they used a transfer-matrix method. In this method, the network is not constructed all at once, but by a step-by-step procedure by adding rows (or planes in 3D) of nodes to the network, starting from the inlet row (or plane). Alvarado *et al.* (1997) used a pore network model similar to that of Koplik *et al.* (1988b), except that they also considered the effect of a first-order reaction and linear adsorption on the dispersion process. Their simulations indicated that if the kinetic coefficients of the reaction and adsorption is the same everywhere, then the dispersion coefficient is the same as that of the nonreactive case. The same is true when the Damköhler number Da is much larger than one. However, when $Da \rightarrow 0$, the description of the dispersion process by a CD equation breaks down.

11.11

Long-Time Tails: Dead-End Pores versus Disorder

As already discussed, molecular diffusion transfers the solute into and out of stagnant, dead-end or low-velocity regions of a pore space. Many measurements of the

concentration profiles of solutes indicate the presence of a long-time tail in the profiles. Diffusion into and out of the stagnant regions is often invoked to explain such long-time tails, which have been of great interest for a long time. Carberry and Bretton (1958), Aris (1959), and Turner (1959) were probably the first who studied dispersion in systems with stagnant regions. In particular, Aris (1959) showed that $D_L/D_m \sim Pe^2$, a result that was rediscovered by Koch and Brady (1985).

In the early 1960s, there were several studies of the relation between the long-time tails and the effect of dead-end pores. Deans (1963) and Coats and Smith (1964) attributed the long-time tails to the presence of dead-end pores that can cause long delays in the travel times of a solute and, hence long tails in its concentration profile. They developed a semi-empirical model to account for the effect, which is described below. Brigham (1974) and Baker (1977) found that trapping in dead-end pores is needed to describe dispersion in carbonate rocks, but not in sandstones. They proposed that the origin of stagnant regions in carbonate rocks is either regular or bimodal porosity. Their proposal was disputed by Gist *et al.* (1990), who measured dispersion coefficients in a variety of sandstones and carbonate porous media. Their mercury capillary-pressure data for the Austin Chalk and Indiana limestone indicated the presence of bimodal porosity, yet no long-time tails were observed in the measured concentration profiles.

Deans (1963), Coats and Smith (1964), Passioura (1971), Baker (1977), Rao *et al.* (1980), and Salter and Mohanty (1982) all investigated the effect of the long-time tails and dead-end pores. In Baker's model, which is the most sophisticated of such works, it is assumed that a fraction ϕ_f of the pore volume is available for flow, while $1-\phi_f$ is the fraction of the stagnant or dead-end fraction. A 1D CD equation is used, modified to account for the effect of the stagnant regions:

$$\phi_f \frac{\partial C_f}{\partial t} + (1 - \phi_f) \frac{\partial C_s}{\partial t} + V \frac{\partial C_f}{\partial x} = D_L \frac{\partial^2 C_f}{\partial x^2}, \quad (11.73)$$

where C_f and C_s are, respectively, the concentrations of the solute in the flowing and stagnant regions. Equation (11.73) is augmented by a mass balance between the stagnant and flowing fluids:

$$(1 - \phi_f) \frac{\partial C_s}{\partial t} = k_c (C_f - C_s), \quad (11.74)$$

where k_c is the mass transfer coefficient such that k_c^{-1} is the time the solute spends in the stagnant regions. Equations (11.73) and (11.74), with the appropriate initial and boundary conditions, are then solved. Normally, ϕ_f and k_c are not known *a priori* and are treated as adjustable parameters.

Bacri *et al.* (1990a) who used an acoustic technique to measure the concentration and velocity profiles during dispersion in unsaturated porous media (i.e., dispersion in one fluid phase in a presence of another immiscible fluid), Charlaix *et al.* (1987b) who measured the dispersion coefficients and concentration profiles in sintered-glass bead packs, and Gist *et al.* (1990) who did the same in a variety of sandstones and carbonate rocks, all used the Coats–Smith–Baker model to fit their

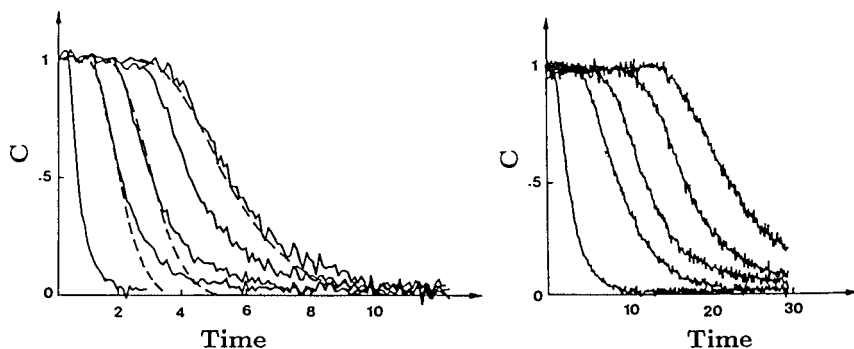


Figure 11.7 Unsaturated concentration profiles C at several cross sections of the porous medium with mean flow velocity $V = 3.6$ cm/h (a) and $V = 0.9$ cm/h (b). The dashed curves correspond to Gaussian profiles (after Bacri *et al.*, 1990a).

data and obtained very accurate fits. However, whereas Bacri *et al.* (1990a) attributed the long-time tails in their data to the length of their porous medium as being too short to allow for the development of the Gaussian dispersion (see Figure 11.7), Charlaix *et al.* (1987b) and Gist *et al.* (1990) attributed them to the heterogeneity of their porous media. Thus, it is important to understand why the Coats–Smith–Baker model is able to provide such good fits to the data.

Based on their studies of dispersion in consolidated porous media, Gist *et al.* (1990) identified two cases in which the long-time tails can occur. The first is that of a heterogeneous porous medium in which the permeability contrast between various regions is strong enough. The heterogeneity gives rise to a long-time tail in the concentration profile. The second case is that of a *narrow* pore-size distribution in which the permeability heterogeneities are due to defects in the packing density. If the long-time tails are in fact due to the permeability heterogeneities, the implications for the upscaling of laboratory results to field conditions are important.

For example, the Coats–Smith–Baker model predicts that the long-time tails will disappear if k_c^{-1} is much smaller than the total travel time of the solute particles (this is easily seen by inspecting Eqs. (11.73) and (11.74)), whereas the long-time tails will persist if there are strong permeability heterogeneities at any length scale. This is also consistent with the studies and measurements of tracer dispersion in groundwater flow in heterogeneous aquifers (Pickens and Grisak, 1981). However, before we go on and explain this complex phenomenon, let us first study dispersion in short porous media, a closely related subject.

11.12

Dispersion in Short Porous Media

We already mentioned that Bacri *et al.* (1990a) attributed the long-time tails in the concentration profiles to the small size of their porous medium. Thus, dispersion

in short porous media can be important because, then, the mixing zone will be large compared with the medium's length. Brenner (1962) and Brigham (1974) were among the first to study the issue, with Brigham (1974) making a comprehensive and definitive analysis of this problem.

We already presented the solution of a CD equation for dispersion in a 1D porous medium of length L and a step change in the inlet concentration at time $t = 0$ (see Eqs. (11.10)) and (11.11). We define

$$\beta_{\pm} = \frac{L \pm V \left(t - \frac{V_{\text{inj}}}{Q} \right)}{\sqrt{4D_L \left(t - \frac{V_{\text{inj}}}{Q} \right)}}, \quad (11.75)$$

where Q is the volume flow rate, and V_{inj} the total volume of the injected tracer solution. Then, the solution for a pulse input of total volume V_{inj} is obtained by superimposing two solutions for step changes, in which case the outlet solute concentration is given by

$$\begin{aligned} \frac{C}{C_0} = & \frac{1}{2} \text{erfc}(\alpha_-) + \frac{1}{2} \exp\left(\frac{LV}{D_L}\right) \text{erfc}(\alpha_+) - \frac{1}{2} \text{erfc}(\beta_-) \\ & - \frac{1}{2} \exp\left(\frac{LV}{D_L}\right) \text{erfc}(\beta_+), \end{aligned} \quad (11.76)$$

where $\alpha_{\pm}$ are defined by Eq. (11.10). If one is only interested in observation times $t \gg V_{\text{inj}}/Q$, then Eq. (11.76) is simplified to

$$\frac{C}{C_0} = \left(\frac{L}{2\pi}\right) \left\{ \left[D_L \left(t - \frac{V_{\text{inj}}}{Q} \right) \right]^{-\frac{1}{2}} - (D_L t)^{-\frac{1}{2}} \right\} [\exp(-\beta_-^2) + \exp(-\alpha_-^2)]. \quad (11.77)$$

Gist *et al.* (1990) used Eq. (11.77) to fit the concentration profiles that they had measured, and found that the resulting fits are as accurate as those provided by the Coats–Smith–Baker model.

Brigham (1974) showed that if the Coats–Smith–Baker model is adjusted at the effluent boundary to account for the difference between the in-situ and flowing concentrations, then Eqs. (11.11), (11.76) and (11.77) and the Coats–Smith–Baker model will essentially provide identical fits to the data. This explains why Bacri *et al.* (1990a) could fit their data for dispersion in a short porous medium with the Coats–Smith–Baker model.

Koch and Brady (1987) also considered dispersion in porous media of short to moderate lengths. They derived an expression for the Fourier transform of the concentration and the effective dispersion coefficients, and showed that the characteristic time τ_{KB} for reaching a diffusive transport described by a CD equation is related to a Péclet number Pe_1 by

$$\tau_{\text{KB}} \sim \text{Pe}_1^{-\frac{2}{3}}, \quad (11.78)$$

where $Pe_1 = dV/D_m$, with d being the typical grain size *before* the grains are fused to produce a consolidated porous medium. Koch and Brady (1987) also found qualitative agreement between Eq. (11.78) and the data of Charlaix *et al.* (1987b). Bacri *et al.* (1990a) also used the Koch–Brady expression for the concentration profile, but found only qualitative agreement between the predictions and their data, whereas the Coats–Smith–Baker model provided an accurate fit to their data. The reason for the discrepancy is perhaps that the Koch–Brady results are valid in the limit of high porosities, whereas the data of Bacri *et al.* (1990a) and Charlaix *et al.* (1987b) were both for porosities that are beyond the region of validity of the Koch–Brady results. Koch and Brady (1987) also proposed that τ_{KB} is the same as k_c^{-1} in the Coats–Smith–Baker model.

11.13

Dispersion in Porous Media with Percolation Disorder

In this section, we describe dispersion in porous media with percolation disorder, modeled by pore networks in which a fraction of the pores (bonds) are cut at random. As Katz and Thompson (1986, 1987) showed (see Chapter 10), flow in a porous medium with a broad pore size or permeability distribution may be mapped onto an equivalent percolation problem. The same must be true about dispersion in such porous media, as a broad pore size distribution gives rise to a broad distribution of pore flow velocities that, in turn, affects dispersion. There are two features of percolation pore networks that influence dispersion. One is the fact that such pore networks have a large number of dead-end pores (bonds) near the percolation threshold p_c (see Chapter 3) and, thus, holdup dispersion may be important. The second feature is that for length scales shorter than the percolation correlation length ξ_p , the sample-spanning cluster and its backbone are fractal objects (see Chapter 3) and, thus, dispersion is not expected to be described by a CD equation. We call this regime fractal dispersion.

To study dispersion in percolation pore networks, two important characteristic quantities are essential: the *dispersivities*, $\alpha_L = D_L/V$, and $\alpha_T = D_T/V$, which are proportional to each other, but α_L is usually larger than α_T . Physically, the dispersivities represent the length scale over which a CD equation can describe dispersion and, thus, in some sense, they are similar to the percolation correlation length ξ_p (see Chapter 3) because dispersion in a percolation network can be described by a CD equation if the dominant length scale of the system is larger than ξ_p .

11.13.1

Theoretical Developments

In their simulations of dispersion in percolation networks, Sahimi *et al.* (1982, 1983a, 1986a,b) found that as p_c is approached, the dispersivities and dispersion coefficients also increase. The increase may be attributed to the fact that near p_c , the transport paths are very tortuous, resulting in broad first-passage time distri-

butions (FPTDs) and, hence, large dispersive mixing of the two fluids. Figure 11.8 shows their results for dispersion in a percolating square network.

We should mention here the work of de Gennes (1983a). He studied dispersion near p_c . After a rather long analysis, he showed that in calculating D_L , the average flow velocity must be based on the *total* travel time of the solute particles in the sample-spanning cluster, rather than the travel time along the backbone *alone*. Intuitively, this is clear. Indeed, in experimental measurements of the concentration profiles and D_L , there is no way of measuring the travel times along the backbone alone. Instead, what is routinely measured is the total travel or transit time.

In Chapter 3, we described universal power laws for various properties of percolation networks near the percolation threshold. What are the power laws for D_L and D_T near p_c ? Our discussions so far must have made it clear that D_L and D_T are sensitive to the structure of a porous medium. Similar to fractal diffusion described in Chapter 10, we may define a crossover time τ_{co} such that for $t \gg \tau_{co}$, dispersion is Gaussian or diffusive and follows a CD equation, whereas for $t \ll \tau_{co}$, dispersion is non-diffusive, with the crossover between the two regimes taking place at about $t \simeq \tau_{co}$. For dispersion near p_c , this time scale is estimated from

$$\tau_{co} \sim \frac{\xi_p^2}{D_L} \quad (11.79)$$

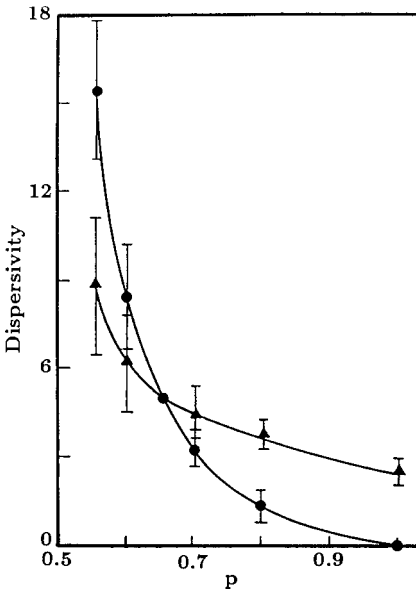


Figure 11.8 Longitudinal (circles) and transverse (triangles) dispersivities in a percolating square network in which a fraction p of the bonds carry flow. The dispersivities are mea-

sured in units of the length of a bond (pore). At $p = 1$, there is no disorder, and thus the dispersion coefficients and the dispersivities are both zero (after Sahimi *et al.*, 1986b).

since the dominant length scale in the system is ξ_p . To derive the power laws for D_L and D_T , we must separately consider the various dispersion regimes described in Section 11.7. Let us first introduce two random-walk fractal dimensions by

$$\langle \Delta x^2 \rangle \sim t^{\frac{2}{D_w^l}}, \quad (11.80)$$

$$\langle y^2 \rangle \sim \langle z^2 \rangle \sim t^{\frac{2}{D_w^t}}, \quad (11.81)$$

where, $\langle \Delta x^2 \rangle = \langle (x - \langle x \rangle)^2 \rangle = \langle x^2 \rangle - \langle x \rangle^2$. Equations (11.80) and (11.81) are defined for length scales $L \ll \xi_p$. Two average flow velocities may also be defined. One is an average velocity V_c , defined in terms of the travel time in the sample-spanning cluster. Then, $V_c \sim K_e/X^A$, where K_e is the effective permeability of the pore network (porous medium), and X^A is the percolation accessible fraction described in Chapter 3. Thus, near p_c ,

$$V_c \sim (p - p_c)^{e-\beta} \sim \xi_p^{-\theta_c}, \quad (11.82)$$

where $\theta_c = (e - \beta)/\nu$, and e , ν and β are, respectively, the critical exponents of the permeability, the correlation length, and the accessible fraction that were defined in Chapter 3. On the other hand, if an average particle velocity V_B is defined in terms of the travel times along the backbone, then $V_B \sim K_e/X^B$, where X^B is the percolation backbone fraction defined in Chapter 3. Thus, near p_c ,

$$V_B \sim (p - p_c)^{e-\beta_B} \sim \xi_p^{-\theta_{bb}}, \quad (11.83)$$

where $\theta_B = (e - \beta_{bb})/\nu$, and β_{bb} is the critical exponent of X^B defined in Chapter 3. For length scales $L \ll \xi_p$, we should replace ξ_p in Eqs. (11.82) and (11.83) by L and, therefore, $V_c \sim L^{-\theta_c}$ and $V_B \sim L^{-\theta_{bb}}$, respectively. We also define a *macroscopic* Péclet number by

$$\text{Pe}_M = \frac{V \xi_p}{D_e}, \quad (11.84)$$

where D_e is the effective diffusivity of the system, and V is either V_c or V_B . For $L \ll \xi_p$, we replace ξ_p in Eq. (11.84) by L . Having defined the essential quantities, we now investigate the power laws that the dispersion coefficients and τ_{co} follow near p_c .

1. Let us first consider the small Péclet number regime described earlier. In this case, convection has no effect and $D_L \sim D_T \sim D \sim (p - p_c)^{\mu_p - \beta}$, as derived in Chapter 3, where μ_p is the critical exponent of the electrical conductivity of the (fluid-saturated) system. For $L \ll \xi_p$, we have fractal diffusion. Based on the discussions in Chapters 9 and 10, we can immediately write

$$D_w^l = D_w^t = D_w = 2 + \theta, \quad (11.85)$$

where $\theta = (\mu_p - \beta)/\nu$ and D_w is the fractal dimension of the random walk defined in Chapter 10. Moreover, according to Eq. (11.79),

$$\tau_{co} \sim (p - p_c)^{-\mu_p - 2\nu + \beta} \sim \xi_p^{2+\theta}, \quad (11.86)$$

so that $\tau_{\text{co}} \sim L^{2+\theta_p}$ for $L \ll \xi_p$. Equations (11.85) are valid for dispersion in the entire sample-spanning cluster in the limit $\text{Pe}_M \rightarrow 0$. For dispersion in the backbone, θ in Eqs. (11.85) and (11.86) should be replaced with $\theta_B = (\mu_p - \beta_{\text{bb}})/\nu$. Moreover, for the n th moment of the FPTD, we have $\langle t^n \rangle \sim \langle t \rangle^n$, where $n > 1$ is an integer and, therefore, for $L \ll \xi_p$, we have $\langle t^n \rangle \sim (L^2/D_e)^n$. Therefore,

$$\langle t^n \rangle \sim L^{n(2+\theta_p)}. \quad (11.87)$$

2. Now, suppose that dispersion only takes place in flow through the backbone of the pore network, and that Pe_M is relatively large. Although any porous medium has a large number of dead-end pores near its percolation threshold (critical porosity) p_c , as the experiments of Charlaix *et al.* (1988a) indicated, the porous medium must be extremely close to p_c if the effect of the dead-end pores is to be seen, so that dispersion along the backbone has practical importance. For dispersion in the backbone, we have $D_L/D_e \sim \text{Pe}_M$, and $D_T/D_e \sim \text{Pe}_M$ (i.e., mechanical dispersion) since the logarithmic correction indicated by Eq. (11.65) is neglected in scaling analyses (as $\ln x$ grows with x slower than any power of x and, therefore, it is equivalent to a zero critical exponent). Therefore, $D_L \sim D_T \sim \xi_p V_B \sim \xi_p^{1-\theta_B} \sim (p - p_c)^{e-\beta_{\text{bb}}-\nu}$. Using the numerical values of e , β_{bb} and ν given in Table 3.2, we obtain $D_L \sim D_T \sim (p - p_c)^{-0.56}$ in 2D, and $D_L \sim D_T \sim (p - p_c)^{0.04}$. Thus, D_L and D_T diverge in 2D, but *vanish* very weakly in 3D. The difference demonstrates the strong effect of the backbone structure on dispersion processes. As described in Chapter 3, the backbone is approximated by nodes, links, and blobs. Links are the bonds or pores that connect the blobs and the remaining multiply-connected bonds aggregate together in the blobs. The blobs are very dense in 2D, providing a wide variety of paths for the solute particles with broad FPTDs. As a result, D_L and D_T both diverge as p_c is approached. On the other hand, the blobs are not dense in 3D, which means that the FPTDs are not broad enough to give rise to divergent D_L and D_T . In a hypothetical porous medium similar to a Bethe lattice (for which $e = 3$, $\beta_{\text{bb}} = 1$ and $\nu = 1/2$), $D_L \sim D_T \sim (p - p_c)^{1/2}$, indicating the strong effect of the closed loops of pores (which are absent in the Bethe lattices) on D_L and D_T .

For $L \ll \xi_p$, we replace ξ_p with L and, thus,

$$D_L \sim L^{1-\theta_B} \quad (11.88)$$

with a similar power law for D_T . As all the length scales of the system must be proportional to each other (and to L), Eq. (11.88) may be rewritten as $D_L \sim \langle \Delta x^2 \rangle^{(1-\theta_B)/2}$. Using a fundamental property of random-walk processes, $D_L \sim d\langle \Delta x^2 \rangle/dt$, we obtain $d\langle \Delta x^2 \rangle/dt \sim \langle \Delta x^2 \rangle^{(1-\theta_B)/2}$, which, after integration, yields

$$\langle \Delta x^2 \rangle \sim t^{\frac{2}{1+\theta_B}}, \quad (11.89)$$

with similar equations for $\langle y^2 \rangle$ and $\langle z^2 \rangle$. Equation (11.89) implies that

$$D_L \sim t^{\frac{1-\theta_B}{1+\theta_B}}, \quad (11.90)$$

and that

$$D_w^l = D_w^t = 1 + \theta_B. \quad (11.91)$$

Equation (11.89) implies that in 2D, $\langle \Delta x^2 \rangle \sim \langle y^2 \rangle \sim t^{1.26}$, and in 3D, $\langle \Delta x^2 \rangle \sim \langle y^2 \rangle \sim \langle z^2 \rangle \sim t^{0.97}$. That is, dispersion is *superdiffusive* in 2D, so that the mean-square displacements of the solute particles grow with time faster than linearly, whereas it is *fractal* or *subdiffusive* in 3D, so that the mean-square displacements grow slower than linearly with time.

On the other hand, according to Eq. (11.83) for $L \ll \xi_p$, we have $V_B \sim L^{-\theta_B} \sim \langle x \rangle^{-\theta_B}$, and since $V_B \sim d\langle x \rangle/dt$, we obtain, after integration,

$$V_B \sim t^{-\frac{\theta_B}{1+\theta_B}}, \quad (11.92)$$

which is in sharp contrast with diffusive dispersion for which V_B is *constant*. Finally, since $\alpha_L = D_L/V_B$, we obtain

$$\alpha_L \sim t^{\frac{1}{1+\theta_B}}, \quad (11.93)$$

which means that for nondiffusive dispersion, the dispersivity depends on t . The time scale τ_{co} is given by

$$\tau_{co} \sim (p - p_c)^{-e+\beta_{bb}-\nu} \sim \xi_p^{1+\theta_B}, \quad (11.94)$$

and for $L \ll \xi_p$, we have $\tau_{co} \sim L^{1+\theta_B}$. Equation (11.94) should be compared with Eq. (11.86). It is not too difficult to show that $\langle t^n \rangle \sim (p - p_c)^{-\nu-n(e-\beta_{bb})} \sim \xi_p^{1+n\theta_B}$, and $\langle t \rangle \sim \xi_p^{1+\theta_B}$. Thus, $\langle t^n \rangle / \langle t \rangle^n \sim \xi_p^{1-n}$. That is, from the scaling of $\langle t \rangle$ alone, one *cannot* obtain the scaling of $\langle t^n \rangle$ for $n > 1$. For $\xi_p \gg L$, we have

$$\langle t^n \rangle \sim L^{1+n\theta_B}, \quad (11.95)$$

which should be compared with Eq. (11.87) if we replace θ by θ_B .

3. Consider the holdup dispersion described earlier. We have $D_L \sim (V_c \xi_p)^2 / D_e$, which is the same as Eq. (11.50) in which the length scale is ξ_p and the molecular diffusivity D_m has been replaced by the effective diffusivity D_e in the porous medium, as suggested by de Gennes (1983b). Therefore, we can write $D_L \sim \xi_p^{2-2\theta_c+\theta} \sim (p - p_c)^{-2\nu-\mu_p-\beta+2e}$, with a similar result for D_T and, thus, $D_L \sim (p - p_c)^{-1.5}$ and $D_L \sim (p - p_c)^{-0.17}$ in 2D and 3D, respectively. That is, as p_c is approached, the dispersion coefficients *diverge*, which undoubtedly is due to the contribution of the dead-end pores and the long times that the solute particles spend there. For $L \ll \xi_p$, we have

$$D_L \sim L^{2-2\theta_c+\theta}. \quad (11.96)$$

Using the same type of reasoning as before, we find that

$$\langle \Delta x^2 \rangle \sim t^{\frac{2}{2\theta_c - \theta}}, \quad (11.97)$$

with similar scalings with the time t for $\langle y^2 \rangle$ and $\langle z^2 \rangle$. Thus,

$$D_L \sim t^{\frac{2-2\theta_c+\theta}{2\theta_c-\theta}}, \quad (11.98)$$

and

$$D_w^l = D_w^t = 2\theta_c - \theta, \quad (11.99)$$

and, therefore, $\langle \Delta x^2 \rangle \sim t^{2.3}$ and $\langle \Delta x^2 \rangle \sim t^{1.1}$ in 2D and 3D, respectively. That is, dispersion is *always superdiffusive* when the holdup dispersion, that is, trapping of the solute particles in the stagnant regions of the pore space, is dominant. It is then straightforward to show that

$$V_c \sim t^{\frac{-\theta_c}{1+\theta_c}}, \quad (11.100)$$

and, therefore,

$$\alpha_L \sim t^{\frac{2+\theta}{(2\theta_c-\theta)(1+\theta_c)}}, \quad (11.101)$$

which is more complex than Eq. (11.93). The time scale τ_{co} is given by

$$\tau_{co} \sim (p - p_c)^{-2e+\mu_p+\beta_p} \sim \xi_p^{2\theta_c-\theta}, \quad (11.102)$$

and $\tau_{co} \sim L^{2\theta_c-\theta}$ for $L \ll \xi_p$. Therefore, we obtain $\langle t^n \rangle \sim (p - p_c)^{-n\nu(\theta+2)+\nu} \sim \xi_p^{n(\theta_p+2)-1}$. Thus, $\langle t^n \rangle / \langle t \rangle^n \sim \xi_p^{n-1}$ and, therefore, the scaling of $\langle t \rangle$ alone is *not* enough for obtaining the scaling of the higher moments $\langle t^n \rangle$ of the FPTD for any $n > 1$. In the $L \ll \xi_p$ regime, we have

$$\langle t^n \rangle \sim L^{n(\theta+2)-1}. \quad (11.103)$$

Aside from Eqs. (11.85)–(11.87), all the above results were derived by Sahimi (1987) and were confirmed by the pore network simulations of Sahimi and Im-dakm (1988) and Koplik *et al.* (1988b). The fact that $\langle t^n \rangle / \langle t \rangle^n$ depends on n means that *there is no unique time scale for characterizing non-Gaussian dispersion* in porous media with percolation disorder. Koplik *et al.* (1988b) also proposed a general scaling equation for the FPTD given by

$$\mathcal{Q}(t) = \frac{1}{t_d} F_s \left(\frac{t}{t_d}, \frac{t_c}{t_d} \right), \quad (11.104)$$

where t_d and t_c are the diffusion and convective time scales, respectively, and $t_d \sim \xi_p^{2+\theta}$, as Eq. (11.86) indicates. t_d is also the largest time that the solute particles

spend in the dead-end pores since the length of the longest dead-end branches is of the order ξ_p . The scaling function F_s has the following limiting behavior

$$F_s(x, \gamma) \rightarrow \begin{cases} F_1(x) & \text{as } \gamma \rightarrow \infty, \\ \gamma F_2(x) & \text{as } \gamma \rightarrow 0. \end{cases} \quad (11.105)$$

The limit of pure diffusion corresponds to $\gamma = t_c/t_d \rightarrow \infty$, whereas the convective limit corresponds to $\gamma \rightarrow 0$. Numerical simulations supported Eq. (11.104).

Related pore network simulations were carried out by Andrade and co-workers (Lee *et al.*, 1999; Makse *et al.*, 2000; Andrade *et al.*, 2000). They utilized pore networks at the percolation threshold and computed several important properties, including the minimum traveling time $t_{\min}$ and the most probable time t^* between two points separated by a distance r , the length $\ell_{\min}$ of the path corresponding to the time $t_{\min}$, and ℓ^* corresponding to t^* . For 2D pore networks at the percolation threshold, they found that $t_{\min} \sim r^{1.33}$, $t^* \sim r^{D_{bb}}$, $\ell_{\min} \sim r^{1.64}$, $\ell^* \sim r^{D_{\min}}$, and $\ell^* \sim r^{D_{op}}$. Here, D_{bb} is the fractal dimension of the backbone (the flow-carrying part of the network; see Chapter 3), $D_{\min}$ is the fractal dimension of the *shortest path* between the two points, and D_{op} is the fractal dimension of the *optimal path* between the two points.

11.13.2

Experimental Measurements

The increase in the dispersivities and the dispersion coefficients near p_c that is shown in Figure 11.8 was confirmed by the experiments of Charlaix *et al.* (1987b, 1988a) and Hulin *et al.* (1988a), who studied dispersion in model porous media and measured D_L . Charlaix *et al.* (1988a) constructed 2D hexagonal networks of pores with diameters that were of the order of millimeters. They reported that as the fraction of the open pores decreased, D_L increased sharply, and that Eq. (11.46) seemed to explain the data. However, even when the dispersion coefficients were measured quite close to p_c , the quadratic dependence of D_L/D_m on Pe , Eq. (11.50), was *not* observed (although the fraction of the dead-end pores is quite large near p_c), presumably because the exchange time between the flowing fluids and the dead-end regions was so long that it could not be detected during the experiment.

Hulin *et al.* (1988a) measured D_L in bidispersed sintered glass materials prepared from mixtures of two sizes of beads. They reported that when the porosity was decreased from 30 to 12% D_L increased by a factor of 30. The results of the two studies also indicated that dispersion is more sensitive to *large-scale* inhomogeneities of a porous medium than to its detailed *local* structure. Somewhat similar results were obtained by Charlaix *et al.* (1987b).

Gist *et al.* (1990) studied dispersion in a variety of sandstones and carbonate rocks, and used the percolation model of Katz and Thompson (1986, 1987), described in Chapter 10, to quantify their results. Following Sahimi *et al.* (1982, 1983a, 1986a,b), they argued that the fundamental quantity to be considered is the ratio ξ_p/d_g , where d_g is the mean grain size. Since $\xi_p/d \sim (X^A)^{-\nu/\beta}$, and as $\nu/\beta \simeq 2$

(see Table 3.2), one can write $\xi_p/d_g \sim (X^A)^{-2}$. Because X^A is roughly proportional to the fluid saturation S , we obtain

$$\frac{\xi_p}{d_g} \sim S^{-2}. \quad (11.106)$$

Gist *et al.* (1990) derived a relation between $\alpha_r = \alpha_L/d_g$ and ξ_p/d_g using the percolation model of Sahimi *et al.* Their final result is

$$\alpha_r \sim \left(\frac{\xi_p}{d_g} \right)^{2.2} \sim S^{-4.4}. \quad (11.107)$$

Gist *et al.*'s data for sandstones, epoxies and carbonates supported the validity of Eq. (11.107) (see Figure 11.9), hence confirming the relevance of percolation-type disorder to dispersion in a heterogeneous porous medium – even when the porous medium does seem to contain such disorder – and the power laws derived above.

The last question to be addressed in this section is: What is the equation for the probability density function $P(\mathbf{r}, t)$ or, equivalently, the (normalized) solute concentration – if dispersion is non-Gaussian? For example, as the discussion of dispersion in percolation networks near p_c indicates, dispersion in a fractal porous medium is not expected to be Gaussian and, thus, $P(\mathbf{r}, t)$ given by Eq. (11.3) is not expected to be valid. In Chapter 10, we discussed the appropriate form of $P(\mathbf{r}, t)$ for diffusion in fractal porous media. For non-Gaussian dispersion in a porous medium with a fractal dimension D_f , Sahimi (1987) proposed the following equation

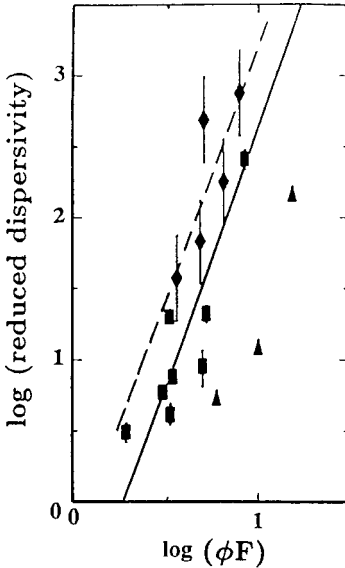


Figure 11.9 Dependence of the reduced longitudinal dispersivity α_L/d_g on the porosity ϕ and the formation factor F in sandstones (squares), epoxies (triangles), and carbonates

(diamonds). Solid and dashed lines represent the best fit of the sandstones and carbonates data, respectively (after Gist *et al.*, 1990).

for $P(r, t)$, in the limit of long times

$$P(r, t) \sim t^{-d_s} \exp \left[-\alpha_1 \left(\frac{|x - \langle x \rangle|}{t^{\frac{1}{D_w^l}}} \right)^{\nu_l} - \alpha_2 \left(\frac{|y|}{t^{\frac{1}{D_w^l}}} \right)^{\nu_t} - \alpha_3 \left(\frac{|z|}{t^{\frac{1}{D_w^l}}} \right)^{\nu_t} \right], \quad (11.108)$$

where α_1 , α_2 and α_3 are constant and, as we already showed, $D_w^l = D_w^t$ for most cases. Here $d_s = D_f/D_w^l$, $\nu_l = D_w^l(D_w^l - 1)^{-1}$, and $\nu_t = D_w^t(D_w^t - 1)^{-1}$. Equation (11.108) reduces to Eq. (11.3) when $D_f = d$ and $D_w^l = D_w^t = 2$. Pore network simulations of Sahimi and Imdakm (1988) appeared to support the validity of Eq. (11.108), but no rigorous derivation of Eq. (11.108) is yet available, and the matter is still an open question.

11.14

Dispersion in Field-Scale Porous Media

Dispersion in field-scale (FS) porous media has attracted considerable attention by hydrologists, petroleum, chemical and environmental engineers, and even *politicians* over the past three decades. The main cause for the attention is the growing concerns about pollution and water quality. Due to the intensifying exploitation of groundwater and the increase in solute concentrations in aquifers caused by salt-water intrusion, leaking repositories, and use of fertilizers, dispersion in the FS porous media has been a main topic of research. Moreover, dispersion in miscible displacement processes is an important phenomenon during oil recovery, and depending on the magnitudes of the dispersion coefficients D_L and D_T and other physical parameters of the process, dispersion may help or hurt the displacement processes and their efficiency.

As discussed in Chapter 1, in studying transport in heterogeneous porous media, one should define precisely what is meant by heterogeneous. Chapter 1 described four important scales of heterogeneities, that is, microscopic, macroscopic, megascopic, and gigascopic scales. Bhattacharya and Gupta (1983) described a variety of length scales, ranging from the kinetic and Taylorian to the Darcy scales, while Dagan (1986) considered length scales, ranging from pore to laboratory to formation to regional levels. Cushman (1984) provided a brief review of the general problem of the development of N -scale transport equations. A complete treatment of the problem at all the relevant length scales is still an open question.

We remind the reader that dispersion in a FS porous medium is purely mechanical, arising as a result of large-scale spatial variations of the permeability of the medium and the resulting random velocity field. Thus, Eqs. (11.48) and (11.49) are generally expected to hold. The length scale used to define the Péclet number Pe in Eqs. (11.48) and (11.49) may be the permeability correlation length ξ_k . In this sense, dispersion in a FS porous medium might seem to be somewhat *simpler* than what is studied so far. One of the main problems to be solved for dispersion in a FS porous medium is the relation between the implied prefactors in Eqs. (11.48)

and (11.49) and the distribution of large-scale heterogeneities of the medium. In addition, it turns out that the dispersion coefficients in the FS porous media may depend on the length scale of the observations and, therefore, the precise scaling of D_L and D_T with the characteristics length(s) is also important.

In the early years of investigating dispersion in the FS porous media with large-scale permeability and porosity variations, a CD equation served as the starting point for analyzing the field data, while the analysis of the problem was based on completely deterministic methods. A considerable amount of data has, however, indicated unequivocally that D_L and D_T measured in a field are larger by several orders of magnitude than those measured in a laboratory, and an entirely deterministic approach cannot provide a completely satisfactory explanation for such data. Moreover, field experiments of Sudicky and Cherry (1979), Pickens and Grisak (1981), Sudicky and Frind (1982), Sudicky *et al.* (1985), and Molz *et al.* (1983) (for reviews see, for example, Gelhar *et al.*, 1992; Vanderborght and Vereecken, 2007) indicate that dispersion coefficients and dispersivities are often scale-dependent, and that the apparent dispersivities α_L and α_T seem to increase with the transit times of the solute particles, similar to those in percolation networks for length scales $L \ll \xi_p$. Figure 11.10, adapted after Arya *et al.* (1988), demonstrates this phenomenon very clearly. As the distance from the source increases, so does the dispersivity with no asymptotic limit in which α_L is constantly being apparent.

Warren and Price (1961) seem to be the first to investigate dispersion in the FS porous media, taking into account the effect of the permeability distribution. They used a Monte Carlo method that will be described later in this chapter. De-

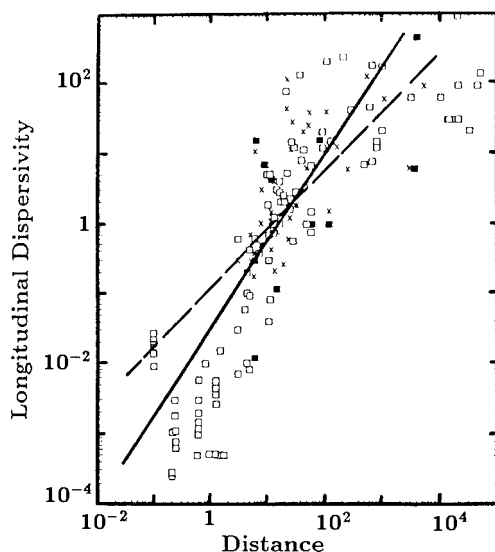


Figure 11.10 Field-scale longitudinal dispersivity as a function of the distance from the source (both are in meters). Solid line represents the best fit to about 75% of the data, while the dashed line represents Gaussian dispersion (after Arya *et al.*, 1988).

spite their many shortcomings, deterministic approaches are still used by many. Although they provide valuable insights into the problem, deterministic approaches fail to provide accurate quantitative predictions for dispersion coefficients and the solute concentration profile. Typical of such approaches is the large-scale volume averaging method of Plumb and Whitaker (1988a,b), which we now describe briefly.

11.14.1

Large-Scale Volume Averaging

Plumb and Whitaker (1988a,b) (see also Thompson and Gray, 1986) considered a *two-scale* problem and developed a large-scale averaging technique for determining the averaged transport equation. The starting point of their analysis was Eq. (11.52), which must be averaged over the regions in which the permeability and porosity vary spatially. To do this, one writes

$$\phi = \{\phi\} + \tilde{\phi}, \quad (11.109)$$

$$\langle \mathbf{v} \rangle = \{\langle \mathbf{v} \rangle\} + \tilde{\mathbf{v}}, \quad (11.110)$$

$$\langle C \rangle = \{\langle C \rangle\} + \tilde{C}, \quad (11.111)$$

$$\mathbf{D}_* = \{\mathbf{D}_*\} + \tilde{\mathbf{D}}, \quad (11.112)$$

where $\{\cdot\}$ denotes the large-scale quantity. By substituting Eqs. (11.109)–(11.112) into Eq. (11.52) and averaging the results over regions in which the permeability and porosity vary appreciably, Plumb and Whitaker (1988a,b) derived a large-scale averaged equation for the average solute concentration that contained such terms as $\nabla \nabla \nabla \{\langle C \rangle\}$ and $\nabla \nabla \partial \{\langle C \rangle\} / \partial t$, indicating that dispersion in the FS porous media cannot, in general, be described by a CD equation.

As our discussion of dispersion in percolation systems in the non-Gaussian regime indicated, when dispersion cannot be described by a CD equation, one must deal with time- and scale-dependent dispersion coefficients. The work of Plumb and Whitaker is valuable in that it clearly demonstrates the deviations of dispersion in the FS porous media from a conventional description by the CD equation. The main difficulty with the approach of Plumb and Whitaker (1988a,b) is that except for very simplified models of a pore space, the numerical solution of their averaged equation is difficult to obtain. The problem must first be solved at the local level in order to use the solution as the starting point for determining the solution for the large-scale averaged equation. Moreover, as discussed earlier, the main contribution to dispersion in a FS porous medium is made by large-scale variations of the permeability and porosity of the medium and local phenomena, such as, diffusion, do not play an important role.

11.14.2

Ensemble Averaging

Koch and Brady (1988) also studied dispersion in the FS porous media, through use of the ensemble-averaging technique described in Chapter 9 and earlier in this chapter. They were able to show that if the correlation length ξ_k for the permeability fluctuations is finite, then dispersion is diffusive and follows a CD equation. If, however, ξ_k is divergent, then superdiffusive dispersion occurs in which the mean-square displacements of the solute particles grow with time faster than linearly, qualitatively similar to superdiffusive dispersion near and at the percolation threshold for length scales $L \ll \xi_p$. In this sense, ξ_k plays a role similar to the percolation correlation length ξ_p . Moreover, Koch and Brady (1988) showed that in the superdiffusive dispersion regime, the space-time evolution of the solute concentration is *universal* and *uniquely* related to the covariance of the permeability field. This is again similar to the superdiffusive dispersion in percolation systems at length scales $L \ll \xi_p$.

11.14.3

Stochastic Spectral Method

Stochastic spectral method has been popular with geologists and hydrologists, and has been used extensively. The main motivation for developing the method is that the complex geohydrological structure of aquifers, the nonuniformity and unsteadiness of flow, and other influencing factors make dispersion in a FS porous medium a very complex phenomenon. Field measurements of the dispersivities are often costly and time consuming. One needs to drill many observation wells to monitor the spread of the solute, and the spreading itself is often so slow that a few *years* may be needed for completing the investigations. The level of uncertainties in all the operations and measurements is quite high and, therefore, stochastic methods have been advocated so that the concepts of randomness, uncertainty and errors can be introduced into the models and analyses.

Early analytical studies of this problem using stochastic concepts were carried out by Mercado (1967) and Buyevich *et al.* (1969), but not all the ingredients were known at that time. Only since the early 1980s has there been a more comprehensive analysis of the problem. The assumption of ergodicity is implicit in the stochastic approaches. That is, one assumes that dispersion of a solute in an ensemble of porous media with the given statistical properties mimics the situation in a real field that is a single realization of a FS porous medium with large-scale variations of the permeability, porosity, and other properties. Such an assumption is valid if the scale of the flow system is large compared with the correlation length of the system. Thus, if the permeability, porosity, or other properties are, for example, fractally distributed (see Chapter 5), that implies that the correlation length may be as large as the linear size of the porous medium, and the stochastic spectral approach may break down.

To give the reader some ideas about stochastic spectral model of dispersion in a FS porous medium, let us describe the work of Gelhar *et al.* (1979), Gelhar and Axness (1983), and Gelhar (1986), which is representative of this class of models (see also Dagan, 1987, and references therein). The starting point is a CD equation at the local level. Assuming, for example, a 2D porous medium (the extension to 3D is described by Gelhar and Axness, 1983), one starts with

$$\frac{\partial C}{\partial t} + \frac{\partial}{\partial x}(vC) = \frac{\partial}{\partial x} \left(D_L \frac{\partial C}{\partial x} \right) + \frac{\partial}{\partial z} \left(D_T \frac{\partial C}{\partial z} \right), \quad (11.113)$$

where it has been assumed that C , D_L and D_T , which are *local* properties, are random processes with

$$C(x, z, t) = C_m(x, t) + c(x, z, t), \quad (11.114)$$

$$v = V_m + u, \quad (11.115)$$

$$D_L = D_{Lm} + d_L, \quad (11.116)$$

$$D_T = D_{Tm} + d_T, \quad (11.117)$$

where subscript m denotes a mean value, for example, $C_m(x, t) = \langle C(x, z, t) \rangle$, with the averaging being taken with respect to the vertical depth z . Here, $c(x, z, t)$, d_L and d_T represent the *fluctuations* with zero averages. If we substitute Eqs. (11.114)–(11.117) into Eq. (11.113) and take the average of both sides, we find that

$$\frac{\partial C_m}{\partial t} + \frac{\partial}{\partial x}(V_m C_m) + \frac{\partial}{\partial x} \langle uc \rangle = \frac{\partial}{\partial x} \left(D_{Lm} \frac{\partial C_m}{\partial x} \right) + \frac{\partial}{\partial x} \left\langle d_L \frac{\partial c}{\partial x} \right\rangle + \frac{\partial}{\partial x} \left\langle d_T \frac{\partial c}{\partial z} \right\rangle, \quad (11.118)$$

where we have used the fact that the averages of the quantities are independent of z . Equation (11.118) is now subtracted from Eq. (11.113), and the coordinate $\xi = x - V_m t$ is used to obtain

$$\begin{aligned} \frac{\partial C}{\partial t} + u \frac{\partial C_m}{\partial \xi} + \frac{\partial}{\partial \xi} (uc - \langle uc \rangle) &= D_{Tm} \frac{\partial^2 c}{\partial z^2} + \frac{\partial}{\partial \xi} \left(d_L \frac{\partial c}{\partial \xi} \right) + D_{Lm} \frac{\partial^2 c}{\partial \xi^2} \\ &+ \frac{\partial}{\partial z} \left(d_T \frac{\partial c}{\partial z} - \left\langle d_T \frac{\partial c}{\partial z} \right\rangle \right) + \frac{\partial}{\partial \xi} \left(d_L \frac{\partial c}{\partial \xi} - \left\langle d_T \frac{\partial c}{\partial \xi} \right\rangle \right). \end{aligned} \quad (11.119)$$

If the perturbation u is small, then the second-order terms (numbered 3, 7 and 8) can be neglected, and one obtains an approximate equation of the form

$$\frac{\partial c}{\partial t} + u \frac{\partial C_m}{\partial \xi} = D_{Tm} \frac{\partial^2 c}{\partial z^2} + d_L \frac{\partial^2 c}{\partial \xi^2} + D_{Lm} \frac{\partial^2 c}{\partial \xi^2}, \quad (11.120)$$

so that even at this level of approximation, one already has the additional term $d_L \partial^2 c / \partial \xi^2$.

Equation (11.120) is solved by assuming that the permeability is a statistically-homogeneous random process, and introducing the spectral representation of the random variables (see Chapter 5). If the field permeability K is written as $K = K_m + k$, where $K_m = \langle K \rangle$, and $\langle k \rangle = 0$, then

$$k = \int_{-\infty}^{\infty} e^{i\omega z} dZ_k(\omega), \quad (11.121)$$

where ω is the wave number and $Z_k(\omega)$ is a complex stochastic process with orthogonal increments. The random processes u , c , d_L , and d_T also have similar spectral representations. Based on the experimental results of Harleman and Rumer (1963) that indicated that α_L seems to be proportional to $\sqrt{K}$ (this may be intuitively clear, as $\sqrt{K}$ is a length scale, as is α_L), it is not difficult to show that $d_L/D_{Lm} = 3k/(2K_m)$. If we introduce a spectral representation for c ,

$$c = \int_{-\infty}^{\infty} e^{i\omega z} dZ_c(\omega), \quad (11.122)$$

then Eq. (11.120) becomes

$$\frac{\partial \gamma}{\partial t} + a_T V_m \omega^2 \gamma - D_{Lm} \frac{\partial^2 \gamma}{\partial \xi^2} = V_s(\xi, t) \quad (11.123)$$

with $\gamma = dZ_c$, and

$$V_s = V_m \frac{dZ_k}{k_m} G, \quad G = -\frac{\partial C_m}{\partial \xi} + \frac{3}{2} a_L \frac{\partial^2 C_m}{\partial \xi^2},$$

and $a_L = D_{Lm}/V_m$, and $a_T = D_{Tm}/V_m$. The cross spectrum of u and c , $S_{uc}(\omega)$, and the spectrum of k , $S_{kk}(\omega)$, are represented by (see, for example, Lumley and Panofsky, 1974)

$$S_{uc}(\omega) = E(dZ_u dZ_c^*), \quad (11.124)$$

$$S_{kk}(\omega) = E(dZ_k dZ_k^*), \quad (11.125)$$

where E denotes the expected value, and $*$ denotes the complex conjugate. Since $dZ_u/V_m = dZ_k/k$, we obtain

$$S_{uc}(\omega) = \frac{S_{kk}(\omega)^2}{k_m^2} \left\{ V_m G \frac{1 - e^{-\beta t}}{a_T \omega^2} - \left(\frac{\partial G}{\partial t} - D_{Lm} \frac{\partial^2 G}{\partial \xi^2} \right) \left[\frac{1 - (1 + \beta t)e^{-\beta t}}{a_T^2 \omega^4} \right] \right\}, \quad (11.126)$$

where $\beta = a_T V_m \omega^2$. Similarly, $\langle uc \rangle$ is given by

$$\langle uc \rangle = A V_m G - B \left(\frac{\partial G}{\partial t} - D_{Lm} \frac{\partial^2 G}{\partial \xi^2} \right), \quad (11.127)$$

and

$$A = \int_{-\infty}^{\infty} \frac{S_{kk}(\omega)}{k_m^2} \frac{1 - e^{-\beta t}}{a_T \omega^2} d\omega, \quad B = \int_{-\infty}^{\infty} \frac{S_{kk}(\omega)}{k_m^2} \frac{1 - e^{-\beta t}}{a_T^2 \omega^4} d\omega.$$

Using all the results, Eq. (11.119) is rewritten as

$$\begin{aligned} \frac{\partial C_m}{\partial t} = & (A + a_L) V_m \frac{\partial^2 C_m}{\partial \zeta^2} - B \frac{\partial^3 C_m}{\partial \zeta^2 \partial t} - 3a_L A V_m \frac{\partial^3 C_m}{\partial \zeta^3} + 3a_L B \frac{\partial^4 C_m}{\partial \zeta^3 \partial t} \\ & + \left(a_L B V_m + \frac{9}{4} a_L^2 A V_m \right) \frac{\partial^4 C_m}{\partial \zeta^4} + \dots \end{aligned} \quad (11.128)$$

Equation (11.128) indicates that the average concentration C_m does not follow a CD equation, a result similar to that obtained by Plumb and Whitaker (1988b). The rest of the analysis is clear: a spectrum $S_{kk}(\omega)$ is assumed and the quantities A and B are calculated. Having determined A and B , one proceeds to analyze Eq. (11.128). Gelhar and Axness (1983) extended the above analysis to 3D heterogeneous media and showed that the dispersion coefficients depend *linearly* on the average velocity, which is not surprising. The results of the stochastic model have been used with some success for predicting the breakthrough of solutes at field scales (see, for example, Kapoor and Gelhar, 1994, and references therein).

The analysis so far assumes that all the randomness in a porous medium is due to the variations of the permeability. An alternative approach relies on a stochastic representation of the velocity (Tang *et al.*, 1982) and develops an ensemble-averaged equation containing coupling between the velocity and the concentration fluctuations (which is similar to that found by Gelhar *et al.*, 1979) that leads to a coefficient in the stochastic transport equation similar to D_L in a conventional CD equation. This term, the *ensemble dispersion coefficient*, depends upon the variance-covariance structure of the velocity field. If the neighboring velocities are uncorrelated, the ensemble dispersion coefficients *increase* as a function of the travel distance from the source. If the covariance of the velocity field is an exponentially-decaying function, then the ensemble dispersion coefficients reach a constant value. Thus, this type of analysis clearly demonstrates the effect of the correlations in the stochastic analysis of dispersion.

The stochastic models of the type that we described here are useful if an adequate representation of the velocity or permeability fields is available. Such representations were described in Chapter 5. The interested reader should consult Dagan (1986, 1987) and Haldorsen and Damsleth (1990) for more details and references on stochastic modeling of transport in the FS porous media.

11.14.4

Continuous-Time Random Walk Approach

In addition to the various theoretical developments described in the last three subsections, there is also considerable experimental evidence that the CD equation

cannot describe dispersion in flow through a large class of heterogeneous porous media. In fact, over 50 years ago, Aronofsky and Heller (1957) had already reported on the deviations of their data for dispersion from the description by a CD equation and noted that the deviations were systematic. Scheidegger (1959) also reported on some careful experiments on dispersion in columns. He noted that the breakthrough solute concentration profile (when the solute first exits the column), when fitted to the CD equation, significantly deviates from his data and commented that, “The deviations are systematic which appears to point toward an additional, hitherto unknown, effect.” Silliman and Simpson (1987) reported convincing data for dispersion in laboratory experiments that indicated scale-dependence of the dispersivity, the hallmark of non-Fickian dispersion.

Such deviations have motivated the development of new models for describing dispersion in flow through heterogeneous porous media. One such model is based on continuous-time random walks (CTRW), an idea whose origin goes back to an important paper of Montroll⁶ and Weiss (1965), and further developed by Montroll and Scher (1973) and Scher and Montroll (1975). The application of the CTRWs to describing dispersion in flow through heterogeneous porous media has been developed by Berkowitz, Cortis, Scher and their co-workers. In this section we describe the basic ideas of the model by closely following the comprehensive review of Berkowitz *et al.* (2006); see also Berkowitz *et al.* (2008)

The starting point of the model is the *master equation* (ME), already used in Chapter 10 for describing diffusion and conduction in porous media. Recall that the ME is given by

$$\frac{\partial C(s, t)}{\partial t} = \sum_{s'} W(s, s') C(s', t) - \sum_{s'} W(s', s) C(s, t), \quad (11.129)$$

where, as in Chapter 10, $C(s, t)$ is the (solute) concentration, or the probability that a solute particle is at s at time t (if the concentration is suitably normalized), and $W(s, s')$ is the transition rate, or the probability of moving from s to s' . The transition rates describe the effect of the flow velocity on the motion of the solute. Note two differences with the formulation presented in Chapter 10. One is that, unlike the analysis in Chapter 10, the transition rates in Eq. (11.129) are not necessarily symmetric. The second difference is that the points s and s' are not necessarily the sites of a lattice. Note also that the ME does not separate the effects of convection and diffusion into distinct parts.

The crucial aspect of the formulation is specification of the transition rates, which entails detailed knowledge of the heterogeneities of porous media. As the discussions throughout this book should have made it clear thus far, below a length scale ℓ_c , the heterogeneities are unresolved. Thus, over such scales, one must resort to a statistical description of the set $\{W(s, s')\}$ through distribution functions. To do so, one resorts to the ensemble-averaged version of Eq. (11.129) which is given

6) *Elliott Waters Montroll* (1916–1983), Albert Einstein Professor of Physics at the University of Rochester, was a mathematician who made very significant contributions to random walks, phase transitions and traffic flow.

by

$$\begin{aligned} \frac{\partial C_m(s, t)}{\partial t} = & \sum_{s'} \int_0^t \varphi(s - s', t - t') C_m(s', t') dt' \\ & - \sum_{s'} \int_0^t \varphi(s' - s, t - t') C_m(s, t') dt' , \end{aligned} \quad (11.130)$$

where $C_m(s, t)$ is the ensemble-averaged concentration, and the memory function $\varphi(s, t)$ will be specified shortly. Note that Eq. (11.130) is fully compatible with Eq. (10.16), except that symmetry of the transition rates was assumed in the formulation of Chapter 10. As mentioned in Chapter 10, Eq. (11.130) is known as the *generalized master equation* (GME). Moreover, as discussed there, going from Eq. (11.129) to Eq. (11.130) forces the governing equation for the ensemble-averaged concentration to be *nonlocal* and contain *memory*. Use of nonlocal transport equations with memory goes back to the work of Zwanzig (1960). Note that although the function φ depends on time, it is still stationary in space, only depending on the difference $s - s'$.

A CTRW is described by

$$P(s, t) = \sum_{s'} \int_0^t \psi(s - s', t - t') P(s', t') dt' , \quad (11.131)$$

where $P(s, t)$ is the probability *per time* that a random walker has just arrived at position s and time t , and $\psi(s, t)$ is the probability per time for a displacement s with a difference of arrival times of t . The initial condition for $P(s, t)$ is that the walker is at the origin at time zero. Equation (11.131), which was first proposed by Montroll and Weiss (1965), is Markovian in space (no step of the random walk depends on the previous steps), but not in time (the equation contains memory). Its first application to a physical system was suggested by Scher and Lax (1973a). As pointed out in Chapter 10, Kenkre *et al.* (1973) and Shlesinger (1974) showed that the GME is completely equivalent to a CTRW. The correspondence between Eqs. (11.130) and (11.131) is given by

$$C_m(s, t) = \int_0^t \Psi(t - t') P(s, t') dt' . \quad (11.132)$$

Here, $\Psi(t)$ is given by

$$\Psi(t) = 1 - \int_0^t \psi(t') dt' , \quad (11.133)$$

and is the probability for the walker to remain – to *wait* – at a point s ,

$$\psi(t) = \sum_s \psi(s, t) , \quad (11.134)$$

and the function φ is related to the ensemble-averaged concentration in the Laplace transform space by

$$\tilde{\varphi}(s, \lambda) = \frac{\lambda \tilde{\psi}(s, \lambda)}{1 - \tilde{\psi}(\lambda)}, \quad (11.135)$$

where λ denotes the Laplace transform variable conjugate to t .

Equations (11.131)–(11.133) are solved by using Fourier transform (Scher and Lax, 1973a). The solution is

$$C(\omega, \lambda) = \frac{1 - \tilde{\psi}(\lambda)}{\lambda} \frac{1}{1 - A(\omega, \lambda)}, \quad (11.136)$$

where $C(\omega, \lambda)$ and $A(\omega, \lambda)$ are, respectively, the Fourier transforms of $\tilde{C}_m(s, \lambda)$ and $\tilde{\psi}(s, \lambda)$. Equation (11.136) is valid for a lattice of N sites with periodic boundary conditions, in which the sites' positions are given by $s = \sum_{j=1}^3 s_j a_j$ with $s_j = 1, 2, \dots, N$, and a_j being the lattice constants. The components ω_i of ω are given by $\omega_i = 2\pi m_i/N$ with $-(N-1)/2 \leq m_i \leq (N-1)/2$ for odd N . If N is very large, the lattice constant can be arbitrarily small, approaching the continuum limit.

Another important quantity, which has been already mentioned and its use described, is the first passage-time distribution $Q(s, t)$, the probability density of a walker arriving at s at time t for the first time. Introducing the FPTD into the CTRW formulation is necessitated by the fact that Eq. (11.136) gives the solution for a system with periodic boundary conditions, whereas such conditions do not necessarily exist in actual experiments. In addition, as mentioned earlier, the breakthrough curve is equivalent to $Q(s, t)$. Thus, we write Eq. (11.4) in a slightly more general form and in the notation adopted for the CTRW:

$$P(s, t) = \delta_{s,0} \delta(t - 0^+) + \int_0^t Q(s, t') P(0, t - t') dt', \quad (11.137)$$

where the first term on the right side indicates the starting point of the random walker. The solution of Eq. (11.137) in the Laplace transform yields

$$\tilde{Q}(s, \lambda) = \frac{\tilde{P}(s, \lambda) - \delta_{s,0}}{\tilde{P}(0, \lambda)}. \quad (11.138)$$

The breakthrough curve f_{BT} at, say $s_1 = L$, is obtained by summing over all the directions s_2 and s_3 :

$$f_{BT} = \sum_{s_2} \sum_{s_3} Q(s_1 = L, s_2, s_3), \quad (11.139)$$

which is similar to Eq. (11.72). The inverse Laplace and/or Fourier transforms of all the solutions presented so far must be computed numerically, which can be done in a number of ways. Cortis and Berkowitz (2005) developed a collection of easy-to-use MATLAB programs and functions to calculate the temporal and spatial function numerically.

11.14.4.1 Relation between the Transition Rates and the Waiting-Time Distribution

As far as the application of the CTRW to describing solute dispersion in heterogeneous porous media is concerned, identifying the distribution $\psi(s, t)$ and its relation with the transition rates $W(s, s')$ are most crucial. Naturally, $\psi(s, t)$ should reflect the heterogeneity of porous media. In the CTRW formulation, it is assumed that the heterogeneities are reflected in the distribution of the flow velocities that, in turn, represent the heterogeneity in the spatial distribution of the permeabilities. One first writes a ME as a random walk equation in order to obtain a transition length and time distribution

$$\psi_{s,s'}(t) = W(s', s) \exp \left[-t \sum_{s''} W(s'', s) \right]. \quad (11.140)$$

Then, summing over all s' , one obtains

$$\psi_s(t) = \sum_{s'} W(s', s) \exp \left[-t \sum_{s'} W(s', s) \right] = -\frac{dQ_s}{dt}, \quad (11.141)$$

with

$$Q_s \equiv \exp \left[-t \sum_{s'} W(s', s) \right], \quad (11.142)$$

$$\psi(t) = -\frac{d}{dt} [[Q_s]], \quad (11.143)$$

where $[[\cdot]]$ denotes an ensemble average. Calculating the ensemble average Eq. (11.143) in its most general form is, however, difficult.

Hence, consider, first, the case of an ordered system with $W(s', s) = W(s' - s)$, and $W_s = \sum_{s'} W(s', s) = \text{constant}$. Then, $\psi(t) = W_s \exp(-W_s t)$, $\tilde{\psi}(\lambda) = W_s / (W_s + \lambda)$, and

$$\tilde{\varphi}(s, \lambda) = W(s), \quad (11.144)$$

which is similar to Eq. (10.16). Observe that, as the discussions in Chapter 10 indicated, even if the porous medium is not ordered, but an effective (and uniform) transition rate W_e (representing the porous medium) can be determined (Chapter 10 described how W_e is determined by an effective-medium approximation), then Eq. (11.144) is still valid. In the more general case of a disordered medium, one can maintain a fixed value $W(s' - s)$, but have a random spatial distribution of the sites s . Thus, if the site density is N_s (N sites per unit volume), one can write (Scher and Lax, 1973b)

$$[[Q_s]] = \exp \left\{ -N_s \int \{1 - \exp[-W(s)t]\} d^3s \right\}. \quad (11.145)$$

A standard transition rate is given by

$$W(s' - s) = W_m \exp \left(-\frac{|s' - s|}{r_0} \right), \quad (11.146)$$

with r_0 and W_m being the range and maximum value of the transition rates. Equation (11.144) can then be evaluated, (Cortis *et al.*, 2004a)

$$\frac{\psi(\tau)}{W_m} = \zeta {}_3F_3 \left[\begin{matrix} 1, & 1, & 1, \\ 2, & 2, & 2, \end{matrix} ; -\tau \right] \exp(-\zeta \tau {}_4F_4) . \quad (11.147)$$

$\tau \equiv W_m t$, $\zeta \equiv 4\pi N_s r_0^3$, and ${}_pF_q$ is the generalized hypergeometric function⁷⁾ (Abramowitz and Stegun, 1970). In Eq. (11.147), the arguments of ${}_4F_4$ in the exponential term is the same as that of ${}_3F_3$ given above. A plot of $\psi(\tau)/W_m$ versus τ indicates that for low values of τ , $\psi(\tau)$ decreases with τ slowly, as in a power law. Thus, for such values of τ , one has

$$\psi(\tau) \sim \tau^{-1-\alpha} , \quad (11.148)$$

which has been used frequently in the application of the CTRW.

The range of the transition rates that are given by Eq. (11.146) depends on the interplay between the spatial extent r_0 and the average separation distance r_N between the sites, where $r_N^{-3} \equiv 4\pi N_s/3$. Note that $\zeta = 3(r_0/r_N)^3$ for $r_0 \ll r_N$. For every ζ , there exists a τ_c such that $\alpha > 2$ for $\tau > \tau_c$, and dispersion evolves toward the Gaussian (Fickian) regime. As such, α and its value may be viewed as describing the asymptotic behavior of $\psi(t)$ over a time range that corresponds to the duration of the measurements or observations. Scher and Lax (1973b) gave the following expression for the large τ limit:

$$\begin{aligned} \frac{\psi(\tau)}{W_m} &= \zeta [(\ln a_1 \tau)^2 + a_2] \\ &+ \tau^{-1} \exp \left\{ -\frac{1}{3} \zeta [(\ln a_1 \tau)^3 + 3a_2 \ln(a_1 \tau) + 2a_3] \right\} , \end{aligned} \quad (11.149)$$

with a_1 , a_2 , and a_3 being three constants.

11.14.4.2 Continuum Limit of the CTRW

Engineers are most familiar with the continuum mechanics and partial differential equations, the best known example of which are the Navier–Stokes and the CD equations. Thus, it is of importance to recognize that the CTRW may be extended to the continuum limit. If the moments of $\psi(s, t)$ exist, then one can expand $C_m(s', \gamma)$ as a Taylor expansion over the finite range of the transition rates:

$$C_m(s', t) \approx C_m(s, t) + (s' - s) \cdot \nabla C_m(s, t) + \frac{1}{2} (s' - s)(s' - s) : \nabla \nabla C_m(s, t) , \quad (11.150)$$

- 7) The generalized hypergeometric function ${}_pF_q$ is a *hypergeometric series*, $\sum_{k=0}^{\infty} c_k x^k$, for which

$$\frac{c_{k+1}}{c_k} = \frac{(k+a_1)(k+a_2)\cdots(k+a_p)}{(k+b_1)(k+b_2)\cdots(k+b_q)(k+1)} .$$

Then, the function is given by

$$\begin{aligned} {}_pF_q \left[\begin{matrix} a_1, & a_2, & \dots, & a_p, \\ b_1, & b_2, & \dots, & b_q, \end{matrix} ; -x \right] \\ = \sum_{k=0}^{\infty} \frac{(a_1)_k (a_2)_k \cdots (a_p)_k}{(b_1)_k (b_2)_k \cdots (b_q)_k} \frac{x^k}{k!} \end{aligned}$$

with $(a)_k = \Gamma(a_k)/\Gamma(a)$.

where $:$ indicates a tensor product. Inserting Eq. (11.150) into Eq. (11.130) yields (Berkowitz *et al.*, 2002; Dentz *et al.*, 2004) the following equation in the Laplace transform space

$$\lambda \tilde{C}_m(s, \lambda) - C_m(s, 0) = -\mathbf{v}_*(\lambda) \cdot \nabla \tilde{C}_m(s, \lambda) + \mathbf{D}_*(\lambda) : \nabla \nabla \tilde{C}_m(s, \lambda), \quad (11.151)$$

where the convective part of the equation is identified with an effective flow velocity,

$$\mathbf{v}_*(\lambda) \equiv \int s \tilde{\varphi}(s, \lambda) d^d s, \quad (11.152)$$

whereas the effective dispersion (diffusion) tensor is given by

$$\mathbf{D}_*(\lambda) \equiv \int \frac{1}{2} s s \tilde{\varphi}(s, \lambda) d^d s. \quad (11.153)$$

The fact that both $\mathbf{v}_*(\lambda)$ and $\mathbf{D}_*(\lambda)$ depend on λ (and, hence, the time t) is a manifestation of the nonlocality of the governing equation (which was also emphasized in Chapter 10). An expansion similar to Eq. (11.150) for a *single realization* (not ensemble-averaged), together with a Taylor expansion of the transition rates $W(s, s')$, yields

$$\frac{\partial C(s, t)}{\partial t} + \mathbf{v}(s) \cdot \nabla C(s, t) = \nabla \cdot \nabla [\mathbf{D}(s) C(s, t)], \quad (11.154)$$

which represents a slight generalization of the CD equation.

In some cases, one can write $\psi(s, t) = p(s)\psi(t)$, where $p(s)$ and $\psi(t)$ are the probability distributions for the length of the jump of the random walker and the waiting time before the jump is made, respectively. The decoupling is justified if the flow velocity correlates very weakly (or not at all) with $|s|$. In many situations, that is indeed the case (see Berkowitz *et al.*, 2006 for more discussions). In this case (Berkowitz *et al.*, 2002; Cortis *et al.*, 2004b; Dentz *et al.*, 2004),

$$\mathbf{v}_*(\lambda) = \tilde{M}(\lambda) \mathbf{v}_\psi, \quad (11.155)$$

$$\mathbf{D}_*(\lambda) = \tilde{M}(\lambda) \mathbf{D}_\psi, \quad (11.156)$$

with a memory function

$$\tilde{M} \equiv t_c \lambda \frac{\tilde{\psi}(\lambda)}{1 - \tilde{\psi}(\lambda)}, \quad (11.157)$$

and

$$\mathbf{v}_\psi = \frac{1}{t_c} \int s p(s) d^d s, \quad (11.158)$$

$$\mathbf{D}_\psi = \frac{1}{t_c} \int \frac{1}{2} s s p(s) d^d s, \quad (11.159)$$

with t_c being a characteristic time. One may also write

$$\mathbf{D}_\psi = a_\psi |\mathbf{v}_\psi|, \quad (11.160)$$

where the dispersivity-like quantity a_ψ is given by

$$a_\psi = \frac{\int \frac{1}{2} \mathbf{s} \mathbf{s} p(\mathbf{s}) d^d \mathbf{s}}{\left| \int \mathbf{s} p(\mathbf{s}) d^d \mathbf{s} \right|}. \quad (11.161)$$

The working transport equation for the ensemble-averaged concentration is then given by

$$\lambda \tilde{C}_m(\mathbf{s}, \lambda) - C_m(\mathbf{s}, 0) = -\tilde{M}(\lambda) [\mathbf{v}_\psi \cdot \nabla \tilde{C}_m(\mathbf{s}, \lambda) - \mathbf{D}_\psi : \nabla \nabla \tilde{C}_m(\mathbf{s}, \lambda)]. \quad (11.162)$$

Note that the transport velocity $\mathbf{v}_\psi$ is *not* the same as the average fluid velocity $\mathbf{v}$, and similarly for the dispersion tensor $\mathbf{D}_\psi$. The mass flux $\mathbf{J}$, defined through the usual relation, $\partial C_m / \partial t = -\nabla \cdot \mathbf{J}$, is then given by

$$\tilde{\mathbf{J}}(\mathbf{s}, \lambda) \equiv \tilde{M}(\lambda) [\mathbf{v}_\psi \tilde{C}_m(\mathbf{s}, \lambda) - \mathbf{D}_\psi \cdot \nabla \tilde{C}_m(\mathbf{s}, \lambda)]. \quad (11.163)$$

Berkowitz *et al.* (2006) presented some asymptotic solution of the governing transport equation for the ensemble-averaged solute concentration. We note that Bijeljic and Blunt (2006) showed that pore-network simulation of dispersion can produce results that are consistent with the CTRW predictions.

11.14.4.3 Application to Laboratory Experiments

Berkowitz *et al.* (2006) reviewed many applications of the CTRW approach to modeling dispersion in laboratory-scale porous media. Here, we consider two of such applications. One is to the classical experiments of Scheidegger (1959) mentioned at the beginning of this section, reported by Cortis and Berkowitz (2004). Scheidegger's experiments were carried out in Berea sandstone cores 30 in long with a diameter of 2 in, and porosity of 0.204. The flux was $1.73 \text{ cm}^3/\text{min}$. The cores were first fully saturated with the solute tracers and subsequently flushed with clear liquid. Schedegger measured the breakthrough curves. Figure 11.11 presents the fit of the data by both a CD equation and the CTRW formulation. Equation (11.148) was used and the most accurate fit was obtained with $\alpha = 1.59$. As Figure 11.11 indicates, the CTRW formulation provides an excellent fit of the data, whereas the CD equation systematically fails to capture the trends in the data.

The second application is to the data of Nielsen and Biggar (1962), who reported breakthrough curves for both saturated and unsaturated porous media. The columns were filled with Aiken clay loam 0.23–0.5 mm aggregates in saturated conditions. Two breakthrough curves for the saturated case were measured with imposed fluid velocity of 3.4 and 0.058 cm/h. Cortis and Berkowitz (2004) reanalyzed the data and fitted them with the CTRW model. Figure 11.12 presents the fit of the data for one of the breakthrough curves using the CTRW formulation, and compares the results with those obtained with the CD equation. Once again, Eq. (11.148) was utilized, and the most accurate fit was obtained with $\alpha = 1.29$. Clearly, the parameter α is not, and cannot be, universal and must be evaluated for each porous medium.

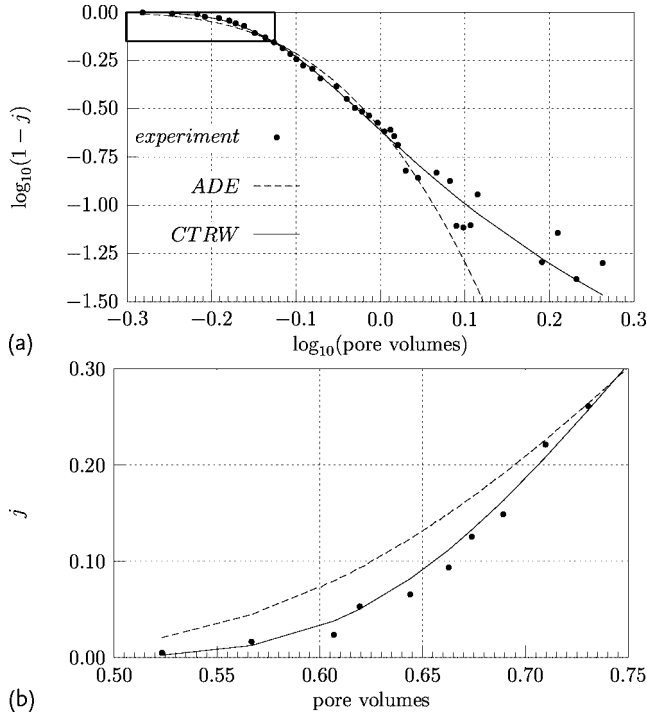


Figure 11.11 The breakthrough data of Scheidegger (1959) in terms of the flux j versus the injected pore volume, and their fit with the convective (advective)-diffusion equation and

the CTRW model. Parts (a) and (b) depict two different plots of the same data (after Cortis and Berkowitz, 2004; courtesy of Professor Brian Berkowitz).

Berkowitz and Scher (2009) discussed more recent developments, including the use of the truncated waiting time distribution,

$$\psi(t) \sim (t_1 + t)^{-1-\alpha} \exp\left(-\frac{t}{t_2}\right), \quad (11.164)$$

where, in addition to α , t_1 and t_2 are other adjustable parameters that are evaluated by fitting the breakthrough curves to the model. Modeling of dispersion in correlated velocity fields based on the CTRW formulation was studied by Berkowitz and Scher (2010). Edery *et al.* (2009) extended the CTRW formulation to the case in which, in addition to the transport, a reaction of the type, $A + B \rightarrow C$, also occurs in porous media.

11.14.4.4 Application to Field-Scale Experiments

The CTRW formulation has also been used to model the data for dispersion of solute in a field-scale (FS) porous medium. Berkowitz and Scher (1998) employed the model in order to explain the data for solute transport in a heterogeneous alluvial aquifer at the Columbus Air Force Base (in Mississippi). In the experiments (which were costly and time consuming), a pulse of bromide was injected

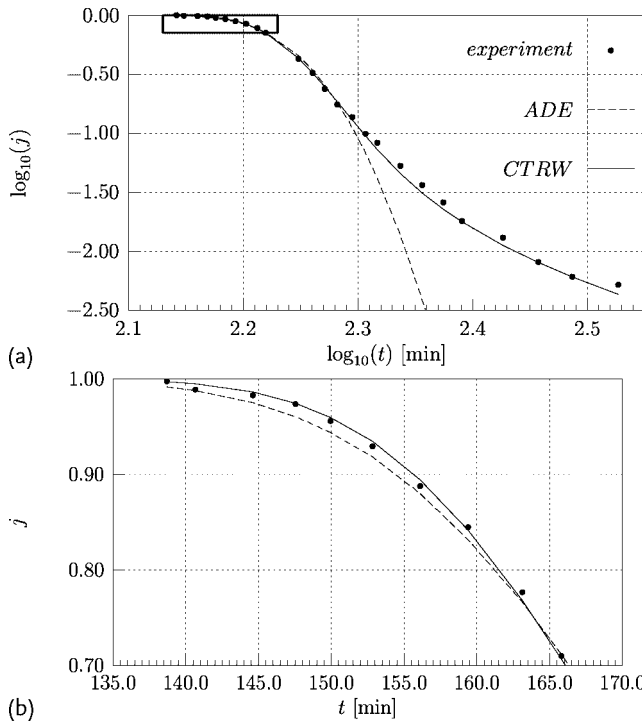


Figure 11.12 The breakthrough data of Nielsen and Biggar (1962) in terms of the flux j versus time t and their fit with the convective (advective)-diffusion equation and the

CTRW model. Parts (a) and (b) depict two different plots of the same data (after Cortis and Berkowitz, 2004; courtesy of Professor Brian Berkowitz).

and traced over a 20 month period. The sampling was done using an extensive 3D well network. The measured tracer plume turned to be completely asymmetric and, hence non-Gaussian. Therefore, the plume could not be described by a CD equation. Berkowitz and Scher utilized the CTRW model to fit the data. The plume shapes obtained by the model displayed the same essential characteristics. In addition, the CTRW model could provide quantitative predictions for the time-dependence of the correlation between the mean and standard deviation of the field plumes and their shape.

In summary, the CTRW formulation is elegant and has yielded considerable insights into non-Fickian transport in heterogeneous porous media. It can also provide quantitative information on many aspects of the transport processes. On the other hand, its parameters, and, in particular, the exponent α defined by Eq. (11.148), are, at this point, purely phenomenological and must be fitted to each and every porous medium. It is not yet clear how to relate the parameters to the fundamental properties of a porous medium and, in particular, its porosity and permeability distribution in the case of the FS porous media, and to the pore size distribution in the case of laboratory-scale porous media.

On the other hand, one may argue that the exponent α is a dynamical property of a porous medium and, thus, it should be related to a directly-measurable dynamical property of the medium. In that case, the leading candidates could be the FPTD and the residence time distribution of the solute particles in a porous medium. However, the problem remains open.

An alternative way is to try to determine an ensemble averaged distribution of the transition rates as defined by Eq. (11.129). If such a distribution can be determined, it will directly lead to the function $\psi(r, t)$ and $\varphi(r, t)$ and, hence, provide clues about the type of functions that one might expect. As the same time, the ensemble-averaged distribution of the transition rates will enable one to directly use the ensemble-averaged master equation to model dispersion in flow through porous media. Progress in this direction has been made; see Cortis *et al.* (2004a,b).

11.14.5

Fractional Convective-Diffusion Equation

Another attempt to model non-Fickian dispersion in flow through heterogeneous porous media has been based on the fractional convective-diffusion (FCD) equation. In Chapter 10, we already briefly described the fractional diffusion (FD) equation for modeling of anomalous diffusion in porous media. In the same spirit, attempts have been made to extend the FD equation to model anomalous dispersion (see, for example, Meerschaert *et al.*, 1999; Benson *et al.*, 2000a,b; Pachepsky *et al.*, 2000; Zhou and Selim, 2003; Deng *et al.*, 2004; Sanchez *et al.*, 2005; Zhang *et al.*, 2005, 2007; Huang *et al.*, 2006; Krepyshcheva *et al.*, 2006). The main motivation for developing this approach has been to develop a formulation that includes two features of non-Fickian dispersion, namely, scale-dependent dispersion coefficients, and the nonlocal nature of the process.

To explain the approach, we consider a 1D porous medium. The generalizations to higher-dimensional porous media have also been developed (Meerschaert *et al.*, 1999). The simplest FCD equation is given by

$$\frac{\partial C}{\partial t} + V \frac{\partial C}{\partial x} = D_L \frac{\partial^\gamma C}{\partial x^\gamma}, \quad (11.165)$$

where the derivative on the right side represents a fractional derivative. Such a derivative can be defined in several ways (see, for example, Samko *et al.*, 1993). One is the Riemann–Liouville definition, given by

$$\frac{\partial^\gamma C}{\partial x^\gamma} = \frac{1}{\Gamma(m - \gamma)} \frac{\partial^m}{\partial x^m} \int_0^x \frac{C(y, t)}{(x - y)^{\gamma - m + 1}} dy, \quad (11.166)$$

which is consistent with, but slightly more general than Eq. (10.60). Here, m is the smallest integer larger than γ , and Γ represents the gamma function. Thus, if $1 < \gamma \leq 2$, then $m = 2$. If $\gamma < 1$, then $m = 1$, in which case we recover Eq. (10.60). The limit $\gamma = 2$ reproduces the conventional CD equation. Equation (11.166) does have some troubling aspects, such as the fact that it yields a nonzero fractional

derivative for a constant value. An alternative definition is the so-called Caputo fractional derivative, defined by

$$\frac{\partial^\gamma C}{\partial x^\gamma} = \frac{1}{\Gamma(2-\gamma)} \int_0^x \frac{1}{(x-y)^{\gamma-1}} \frac{\partial^2 C}{\partial y^2} dy, \quad (11.167)$$

but Eq. (11.166) is often used because it is consistent with the mass conservation equation (see below). Equation (11.166) is also written in an equivalent form (with $m = 2$):

$$\frac{\partial^\gamma C}{\partial x^\gamma} = \frac{\partial}{\partial x} \left[\frac{1}{\Gamma(2-\gamma)} \frac{\partial}{\partial x} \int_0^x \frac{C(y, t)}{(x-y)^{\gamma-1}} dy \right], \quad (11.168)$$

from which it follows that

$$\frac{\partial^\gamma C}{\partial (-x)^\gamma} = \frac{\partial}{\partial x} \left[\frac{1}{\Gamma(2-\gamma)} \frac{\partial}{\partial x} \int_x^L \frac{C(y, t)}{(y-x)^{\gamma-1}} dy \right], \quad (11.169)$$

where L is the length of the 1D system. However, regardless of what definition of a fractional derivative one utilizes, one aspect is clear: any FCD equation is nonlocal because the fractional derivative involves convolution integrals. The same is of course true about the CTRW formulation described in the last section.

Equation (11.165) assumes that the dispersion coefficient D_L does not depend on any length scale (i.e., D_L does not depend on x). However, as discussed earlier, in many cases (depending on the degree of the heterogeneities), scale-dependence of the dispersion coefficient in flow through heterogeneous porous media, even at laboratory scale, has been convincingly established by experiments (Silliman and Simpson, 1987) as well as by pore-scale simulations (Zhang and Lv, 2007). Hence, to include the scale-dependence of D_L in the FCD equation, Eq. (11.165) was generalized, first to (Lu *et al.*, 2002),

$$\frac{\partial C}{\partial t} + \frac{\partial}{\partial x}[V(x)C] = \frac{\partial}{\partial x} \left[D_L(x) \frac{\partial^{\gamma-1} C}{\partial x^{\gamma-1}} \right], \quad (11.170)$$

and then to (Zhang *et al.*, 2006),

$$\frac{\partial C}{\partial t} + \frac{\partial}{\partial x}[V(x)C] = \frac{\partial^{\gamma-1}}{\partial x^{\gamma-1}} \left[D_L(x) \frac{\partial C}{\partial x} \right]. \quad (11.171)$$

What is the form of the dispersive flux j if a FCD equation governs dispersion? Writing down an expression for j entails generalizing Fick's first law. Paradisi *et al.* (2001) suggested that

$$j = -\frac{1}{2} D_L \left[(1+s) \frac{\partial^{\gamma-1} C}{\partial x^{\gamma-1}} + (1-s) \frac{\partial^{\gamma-1} C}{\partial (-x)^{\gamma-1}} \right], \quad (11.172)$$

where $1 \leq s \leq 1$ is the skewness. Equation (11.172) reduces to Fick's first law when $\gamma = 2$. As described earlier in this chapter, one may have a superdiffusive dispersion regime in which some of the solute particles are far ahead of the average motion. This limit corresponds to $s = 1$. In a FS porous medium, superdiffusive dispersion may occur if the spatial distribution of the permeabilities is very broad and contains long-range correlations.

If Eq. (11.172) is used in conjunction with the mass conservation equation, Eq. (11.170) is obtained. On the other hand, if we set $s = 1$ in Eq. (11.172) and replace the first term on its right side by

$$-\frac{\partial^{\gamma-2}}{\partial x^{\gamma-2}} \left[D_L(x) \frac{\partial C}{\partial x} \right],$$

we recover Eq. (11.171). Zhang *et al.* (2007) proposed yet another FCD equation,

$$\frac{\partial C}{\partial t} + \frac{\partial^{\gamma-1}}{\partial x^{\gamma-1}} [V(x)C] = \frac{\partial^{\gamma-1}}{\partial x^{\gamma-1}} \left[D_L(x) \frac{\partial C}{\partial x} \right], \quad (11.173)$$

which implies that the *total* flux J (convective plus dispersive) is given by

$$J = \frac{\partial^{\gamma-2}}{\partial x^{\gamma-2}} \left[V(x)C - D_L(x) \frac{\partial C}{\partial x} \right]. \quad (11.174)$$

Thus, in this formulation, the total flux is also given by a nonlocal equation.

In any case, use of the FCD equation in the modeling of dispersion in FS porous media has had some success; see, for example, Zhang *et al.* (2005). On the other hand, as the discussion makes it clear, there is still some debate as to what the general form of the generalized Fick's law is, and how the nonlocality should be included by the fractional derivatives. The problem is still open.

11.14.6

The Critical Path Analysis

In Chapter 9, we described the critical path analysis (CPA) that has been used for deriving highly accurate expressions for the effective permeability and electrical conductivity of porous media. The CPA was extended by Hunt and Skinner (2008, 2010) in order to derive expressions for the dispersion coefficient D_L and the distribution of the travel times of the solute particles.

Recall that in CPA, transport in a highly heterogeneous porous medium is mapped onto a percolation system at or very near the percolation threshold p_c . Thus, Hunt and Skinner first defined the tortuosity τ_p of the percolation cluster, and in particular that of the backbone – the flow-carrying part of the cluster. Near p_c the length of the shortest path $\xi_{\min}$ on the backbone follows a power law,

$$\xi_{\min} \sim |p - p_c|^{-\eta}, \quad (11.175)$$

where η is a universal exponent. The tortuosity was then defined by Hunt and Skinner as (others have defined τ_p differently) $\tau_p = \xi_{\min}/\xi_p$, where ξ_p is the correlation length of percolation. As described in Chapter 3, near p_c , $\xi_p \sim |p - p_c|^{-\nu}$,

where ν is the associated universal exponent. Hence,

$$\tau_p \sim |p - p_c|^{\nu-\eta}. \quad (11.176)$$

On the other hand, as mentioned earlier, Lee *et al.* (1999) studied the statistics of the travel times of solute particles in the percolation clusters near p_c . They showed that the time t^* for traveling along the most probable path between two points separated by a distance x scales as

$$t^* \sim x^{D_{bb}}, \quad (11.177)$$

where D_{bb} is the fractal dimension of the backbone (see Chapter 3). To derive their results, Hunt and Skinner assumed a particular form for the pore size distribution $f_p(r)$ of the pore space, namely,

$$f_p(r) \propto r^{-D_p-1}, \quad r_0 < r < r_M, \quad (11.178)$$

where D_p is the fractal dimension of the pore space that, as described in Chapters 4 and 9, is not necessarily the same as the fractal dimension of the sample-spanning percolation cluster. Here, r_0 and r_M are, respectively, the minimum and maximum pore sizes. The porosity of the porous medium is then given by

$$\phi = \frac{3-D_p}{r_M^{3-D_p}} \int_{r_0}^{r_M} r^3 f_p(r) dr = 1 - \left(\frac{r_0}{r_M} \right)^{3-D_p}. \quad (11.179)$$

The volume fraction V_p of the pores with sizes $r_0 < r < r_M$ is given by $V_p = \int_{r_0}^{r_M} r^3 f_p(r) dr$. As described in Chapter 9, in the CPA, one identifies a critical pore size r_c as the smallest pore on the sample-spanning cluster that has the largest possible value of the smallest pore. Such a pore controls the flow on the backbone by acting as a bottleneck. Thus, one may define a critical volume fraction V_c corresponding to r_c by $V_c = \int_{r_c}^{r_M} r^3 f_p(r) dr$. In laminar flow through a pore, the flow conductance g is proportional to r^4/l_p , where l_p is the pore's length that is usually taken to be $l_p \sim 1/r$. Hence, $g \sim r^3$ and, therefore,

$$p - p_c \propto \frac{g^{1-\frac{D_p}{3}} - g_c^{1-\frac{D_p}{3}}}{g_c^{1-\frac{D_p}{3}}},$$

where g_c is the critical conductance corresponding to r_c . Hunt and Skinner then used the cluster size distribution n_s of the percolation clusters. As described in Chapter 3, n_s is the number of clusters of size s (normalized by the volume L^d of the system, with d being the dimensionality of the system). Then, consider a path of N resistances along a quasi-1D path of the fluid on the backbone. One has $n_s ds = n_N dN$, where n_N is the number of paths of N resistances. Recall from Chapter 3 that n_s follows a universal scaling equation. More explicitly,

$$n_s \sim s^{-\tau} \exp[-s^{2\sigma}(p - p_c)^2], \quad (11.180)$$

where τ and σ are two percolation exponents (see Table 3.2) such that $\tau - 1 = \sigma \nu d$. Thus, one obtains

$$n_N = N^{-d-1} \exp \left\{ - \left[\left(\frac{Nl}{L} \right)^{\frac{1}{\nu}} \left| 1 - \left(\frac{g}{g_c} \right)^{1-\frac{D_p}{3}} \right| \right]^2 \right\}. \quad (11.181)$$

The probability that a given porous medium of Euclidean length Nl is spanned by a cluster that contains a controlling conductance g is proportional to the integral of $N^d n_N$ over clusters of all sizes larger than or equal to the volume in question. Then, the conductance distribution $f_c(g)$ is given by

$$f_c(g) \propto 2\nu^{-1} Ei \left[\left| 1 - \left(\frac{g}{g_c} \right)^{1-\frac{D_p}{3}} \right|^2 \left(\frac{x}{L} \right)^{\frac{2}{\nu}} \right], \quad (11.182)$$

where x is the linear extent of the porous medium, L now represents the linear size of the representative elementary volume, and Ei is the exponential integral, $Ei(\gamma) = \int_{\gamma}^{\infty} z^{-1} \exp(-z) dz$. An accurate approximation to Eq. (11.182) is

$$f_c(g) \propto \ln \left[\left(\frac{L}{l+x} \right)^{\frac{1}{\nu}} \left| 1 - \left(\frac{g}{g_c} \right)^{1-\frac{D_p}{3}} \right|^{-1} \right]. \quad (11.183)$$

Hunt and Skinner argued that $Q(t)dt = g f_c(g) dg$ since the fluid flux is proportional to $g f_c(g)$. Here, $Q(t)$ is the distribution of the arrival times, or the first-passage time distribution already described. They then assumed that the flow path is quasi-1D, but corrected by the tortuosity τ_p . The time that a solute particle takes to travel a pore is $t \propto rA/q$, where A is the cross-sectional area of the pore and q is the volume flow rate. All the pores along the critical percolation path have the same conductance (see Chapter 9), and $q \propto r^3 t_0$, where t_0 is a pore time scale for flow. Thus, the passage time (without considering the correction due to the tortuosity) is given by

$$\begin{aligned} t(r) &\propto t_0 \int_r^{r_M} \frac{r'^3}{q} \frac{f_p(r')}{r^{-D_p}} dr' \\ &= t_0 \left[\int_r^{r_c} \frac{r'^3}{q} \frac{(r')^{-D_p-1}}{r^{-D_p}} dr' + \int_{r_c}^{r_M} \frac{r'^3}{q} \frac{(r')^{-D_p-1}}{r^{-D_p}} dr' \right]. \end{aligned} \quad (11.184)$$

The tortuosity makes the path longer. Equation (11.176) is rewritten as

$$\tau_p \sim |V_p - V_c|^{\nu-\eta} = |V_p - V_c|^{\nu-\nu D_{bb}}, \quad (11.185)$$

where the relation $\eta = \nu D_{bb}$ was employed. Moreover, Eq. (11.177) also enters the derivation, as it provides t with the explicit dependence on x . Thus, putting

everything together and carrying out the integrations in Eq. (11.184), one finally obtains

$$t = t_0 \left(\frac{x}{L} \right)^{D_{bb}} \frac{1}{3 - D_p} \frac{g_c}{g} \frac{1}{(1 - V_c)^{\eta - \nu}} \left[\frac{1}{1 - V_c} \left(\frac{g_c}{g} \right)^{1 - \frac{D_p}{3}} - 1 \right] \\ \times \left| \left(\frac{g}{g_c} \right)^{1 - \frac{D_p}{3}} - 1 \right|^{\frac{1 - D_{bb}}{\nu}} \equiv t_g \left(\frac{x}{L} \right)^{D_{bb}}. \quad (11.186)$$

One may also determine the distribution $P(x; t)$ of clusters of size of at least x that are dominated by minimum conductances g at a *fixed* time t . Note that x represents both the size of a cluster of connected pores as well as the distance. Then, the dispersion coefficient D_L is simply $D_L = (\langle x^2 \rangle - \langle x \rangle^2)/(2t)$ (for an effectively 1D system). From Eq. (11.177), we obtain

$$x = L \left(\frac{t}{t_g} \right)^{\frac{1}{D_{bb}}}, \quad (11.187)$$

where t_g is defined by Eq. (11.186). Using Eq. (11.183), one obtains

$$f_c(g) \propto \ln \left\{ \left[\frac{L}{l + L \left(\frac{t}{t_g} \right)^{\frac{1}{D_{bb}}}} \right]^{\frac{1}{\nu}} \left| 1 - \left(\frac{g}{g_c} \right)^{1 - D_p} \right|^{-1} \right\}. \quad (11.188)$$

The distribution $P(x; t)$ is then obtained from $P(x; t)dx = f_c(g)dg$, where $f_c(g)$ is given by Eq. (11.188), and x and t are related through Eq. (11.187), with t_g given by Eq. (11.186).

A comparison of the CPA predictions with the experimental data of Nielsen and Biggar (1962), as analyzed by Cortis and Berkowitz (2004), indicated that the predictions are in rough agreement with the data. Thus, clearly, the CPA does have the potential of being able to provide accurate predictions for dispersion in heterogeneous porous media, although it must still be refined.

11.15

Numerical Simulation

Section 11.10 described various models and computer simulation methods for simulating dispersion in laboratory-scale porous media. Before we begin to describe numerical simulation of dispersion in larger-scale, and in particular field-scale (FS) porous media, we should mention that the lattice Boltzmann (LB) method, which was described in Chapter 9 for simulating fluid flow in porous media, has also been extended for simulating dispersion in porous media. Hence, let us first describe the method.

11.15.1

Lattice-Boltzmann Method

Flekkøy (1993) extended the standard LB model based on the Bhatnagar–Gross–Krook (BGK) (Bhatnagar *et al.*, 1954) collision rules (see Chapter 9) in order to simulate diffusion of one component, say B, in another component, say A. Flekkøy *et al.* (1995) extended the model further in order to simulate dispersion in a flowing fluid. Maier *et al.* (1998, 2000) utilized the LB method to simulate dispersion in packed beds, while Jimenez-Hornero *et al.* (2005) used the LB approach to model dispersion in soil.

We assume that component B is present only in trace amounts so that the A–B and B–A collisions can be neglected. Component A has the same equilibrium distribution that was described in Chapter 9, but component B – the solute – evolves towards a new equilibrium as expressed by its own equilibrium distribution function. Since B is present only in small amounts, its equilibrium distribution function contains only up to the first-order term in fluid velocity:

$$n_{Bi}^{\text{eq}}(\mathbf{x}) = w_i \rho_B(\mathbf{x}) (c_s^2 + \mathbf{v}_i \cdot \mathbf{v}_A), \quad (11.189)$$

where w_i is the weight, ρ_B is the density of B, c_s is the speed of sound, $\mathbf{v}_A$ is the velocity vector of A, and $\mathbf{v}_i$ is the microscopic velocities. The weights depend on the structure of the grid used in the simulations. For example, with a square grid with diagonal connections (so that each grid point is connected to eight others), the weights are $w_i = 1/3$ for $i = 1-4$ (the four main directions leading to a grid point along the Cartesian axes), $w_i = 1/12$ for $i = 5-8$ (for diagonal links), and $w_0 = 4/9$ for a particle at rest. The density B is computed by the same equation that we use for component A, Eq. (9.141). Note that because B is present in trace amounts, its velocity is assumed to be the same as that of A, $\mathbf{v}_A$. The molecular diffusivity D_m is given by Flekkøy (1993)

$$D_m = c_s^2 \left(\tau_D - \frac{1}{2} \right), \quad (11.190)$$

where τ_D is a relaxation time for diffusion. The dispersion coefficient D_L is still computed from the mean-square displacements of the B particles, namely, $D_L = (\langle x^2 \rangle - \langle x \rangle^2)/(2t)$, where the position of the B particles is evolved according to the LB simulation. The choice of the time step Δt is important, as it must minimize the number of displacements of the particles, but also have enough accuracy. Suppose that l is the distance between two neighboring nodes. Then, Maier *et al.* (2000) suggested that Δt must be selected such that

$$v_{\text{max}} \Delta t + 2\sqrt{D_m \Delta t} \leq \frac{1}{2} l, \quad (11.191)$$

where v_{max} is the maximum velocity.

The LB method as described only yields one dispersion coefficient, D_L . However, as we have emphasized throughout this chapter, the solute spreads anisotropically,

with the anisotropy being characterized by the dispersion coefficients D_L and D_T . Therefore, it is essential to develop an “anisotropic” LB (ALB) model that can also yield D_T . Zhang *et al.* (2002) and Ginzburg (2005) developed such ALB methods. For example, in their 2D simulations using a LB model with four velocities in nine directions (see Chapter 9), Zhang *et al.* (2002) introduced nine relaxation times τ_i , but with the constraint that $\tau_i = \tau_{i+4}$ along with τ_0 . Then, the dispersion coefficients are given by

$$D_{xx} = \frac{\Delta x^2}{18\Delta t} (4\mu_1 + \mu_2 + \mu_3 - 3), \quad (11.192)$$

$$D_{yy} = \frac{\Delta y^2}{18\Delta t} (4\mu_4 + \mu_2 + \mu_3 - 3), \quad (11.193)$$

$$D_{xy} = \frac{\Delta x \Delta y}{18\Delta t} (\mu_2 - \mu_3), \quad (11.194)$$

where, $D_{xx} = D_L$, $D_{yy} = D_T$, $D_{xy} = D_{yx}$ is the off-diagonal dispersion coefficient in the dispersion coefficient tensor, and $\mu_1 = \tau_1/\Delta t$, $\mu_2 = \tau_2/\Delta t$, $\mu_3 = \tau_4/\Delta t$, and $\mu_4 = \tau_3/\Delta t$. The relaxation parameters are selected such that they are close to each other, but not too close to 0.5, in order to ensure stable solutions. If the goal of the study is to determine the evolution with the time and length scale of the solute concentration, the dispersion coefficients are set, and Eqs. (11.192)–(11.194) are used to estimate the relaxation parameters. Since there are three dispersion coefficients but four relaxation parameters, one of the parameters must be set independent of the dispersion coefficient.

Thus, the method cannot yield information on the dispersion coefficients, but may be used for studying the evolution of the solute concentration. Ginzburg (2005)’s method is more rigorous than that of Zhang *et al.*, but it is also much more involved.

11.15.2

Particle-Tracking Method

The LB method is not efficient enough for simulating dispersion in the FS porous media. In addition, a FS porous medium is characterized by spatial distributions of the permeability and porosity, rather than a pore size distribution. In its present form, the LB model cannot be extended to include the spatial distribution of the permeabilities.

We already described in Chapter 5 models of the FS porous media that were developed by Warren and Price (1961), Warren and Skiba (1964), and Heller (1972), and their improvement by Smith and Freeze (1979) and Smith and Schwartz (1980, 1981a,b), who incorporated short-range correlations between the neighboring blocks. Smith and Schwartz used the model for studying dispersion in the FS porous media. They used an algorithm for the motion of the solute particles that included both deterministic and random displacements. The algorithm is popularly known as the particle-tracking method and is a combination of a random walk and a deterministic component.

In the simulations of Smith and Schwartz, a solute particle is released in the flow field. For each time step Δt , a fluid velocity at any point is calculated by linearly interpolating the four surrounding values (in a 2D system). The particle is then moved a distance that is fixed by the magnitude of the time step and the fluid velocity. This step represents the deterministic portion of the displacement, which is due to convection. Random relocation from the deterministic position, due to dispersion, is accomplished first by moving the particle a distance d_x in a direction that coincides with the flow vector, and second a distance d_y in a direction normal to it. The random displacements d_x and d_y are calculated from

$$d_x = (0.5 - [R])\sqrt{24 D_L^{(l)} \Delta t}, \quad (11.195)$$

$$d_y = (0.5 - [R])\sqrt{24 D_T^{(l)} \Delta t}, \quad (11.196)$$

where $[R]$ a random number uniformly distributed in $(0, 1)$, and $D_L^{(l)}$ and $D_T^{(l)}$ are the *local* dispersion coefficients, that is, the dispersion coefficients on the scale of the size of the grid blocks. Using the model, Smith and Schwartz investigated many aspects of dispersion in heterogeneous porous media and showed that strong permeability heterogeneity gives rise to non-Gaussian dispersion, and that when the permeability correlation length ξ_k is of the order of the system length, a unique dispersion coefficient may not be possible to define, in agreement with the results described earlier. The simulations by Smith and Schwartz represent some of the earliest computational evidence for non-Gaussian dispersion in heterogeneous porous media.

The particle-tracking method is now the standard approach for simulating dispersion in heterogeneous porous media.

11.15.3

Fractal Models

In our discussions of the various variograms in Chapter 5, we mentioned the work of Hewett (1986) (and those of several others) who analyzed the porosity logs of the FS porous media and found that the porosity distributions often follow fractal statistics. More precisely, the vertical porosity logs seem to follow the statistics of the fractional Gaussian noise (FGN), while the lateral logs seemed to be described by the fractional Brownian motion (FBM). Vertical porosity logs analyzed by Hewett (1986) produced values of the Hurst exponent $H \simeq 0.7-0.8$, indicating long-range positive correlations.

Arya *et al.* (1988) analyzed over 130 greatly-varying dispersivities, collected on length scales up to 100 km. The data collected by them, which are shown in Figure 11.10, exhibit large scatter, but their analysis indicated that about 75% of them follow the following scaling law, that is,

$$\alpha_L \sim L^\delta, \quad (11.197)$$

where L is the length scale of measurements, or the distance from the source where the solute had been injected into the solvent in the porous medium. Arya *et al.* (1988) suggested that $\delta \simeq 0.75$. Neuman (1990) presented a different analysis of these and other data, and proposed that there are in fact *two* distinct regimes. One is for $L \ll 100$ m, for which $\delta \simeq 1.5$, whereas the second regime is for $L \gg 100$ m, for which δ was found to be close to unity. Equation (11.197) is reminiscent of dispersion in percolation networks described earlier in this chapter. As the discussions indicated, scale-dependence of α_L implies that it is time dependent as well. Thus,

$$\alpha_L \sim t^v, \quad (11.198)$$

which is again similar to the results for a percolation system at length scales $L \ll \xi_p$ described earlier. A *non-universal* v has been found to provide reasonably accurate fits of various field data, including those shown in Figure 11.10 with $v \simeq 0.5$ – 0.6 . Of course, scale- and time-dependence of α_L implies the same for the fluid velocity V and D_L .

In Hewett's work, the dispersivity α_L was implicitly assumed to follow Eq. (11.198) with

$$v = 2H - 1. \quad (11.199)$$

Therefore, with $H \simeq 0.75$, one obtains $\alpha_L \sim t^{0.5}$. A similar result was obtained by Philip (1986) and Ababou and Gelhar (1990), and was also implicitly assumed by Arya *et al.* (1988). Philip's work also predicted that at short times, $\alpha_L \sim t$, and the constraint $0.5 < H < 1$ was also proposed, consistent with Hewett's analysis of the porosity logs. In a series of paper by the Chevron group, Eq. (11.198) was used in the numerical simulation of flow phenomena in oil reservoirs (Mathews *et al.*, 1989; Emanuel *et al.*, 1989; Hewett and Behrens, 1990). The phenomena that were studied included dispersion, miscible displacements (to be described in Chapter 13), and a waterflood (see Chapter 14).

How are such numerical simulations with fractal distributions of the permeability and/or porosity carried out? Consider, for example, simulation of dispersion. One first generates the permeability and porosity fields using the FBM and FGN distributions. Methods for synthetic generations of the FBM and FGN were described in Chapter 5. The simulations are conditional (see Chapter 5) because the permeability and porosity fields must honor the actual data collected at certain places in the field. The flow field is calculated using Darcy's law, which reduces the problem to calculation of the pressure field. Often, a finite-difference approximation with a rectangular grid is used. The CD equation is then discretized and solved with the resulting flow field. Thus, the implicit assumption is that at the scale of the grid blocks, the CD equation governs dispersion. The assumption is justified, as each block is assumed to be homogeneous. In practice, the discretized equations must be solved for a finely-structured grid so that the effect of the permeability and porosity distributions and their long-range correlations are captured. Comparison of the simulation results with the field data indicates that such conditional simulations with fractal distributions of the permeability and porosity are far

more realistic than the conventional simulations without such distributions. They also demonstrate most definitively the relevance of fractal statistics to modeling the FS porous media and transport processes in them.

11.15.4

Long-Range Correlated Percolation Model

The percolation models that we have described and used so far to model and explain various phenomena in porous media represent *random* percolation models. Such models provide us with a relatively simple theoretical foundation for the existence of scale- and time-dependent dispersion coefficients and dispersivities in the FS porous media. Random percolation models, however, predict *universal* scaling of α_L and D_L with L and t , whereas, as discussed above, field measurements and observations indicate nonuniversal scalings. We now describe a correlated percolation model based on a FBM distribution of the permeabilities, which we utilize to explain the scale-dependence of the dispersivities and the dispersion coefficients of the FS porous media described above. For more details, see Sahimi (1994b).

To each bond of a network, we assign a number selected from a FBM, and interpret it as the effective permeability of a portion of the porous medium over which it is homogeneous. The permeabilities are, therefore, infinitely correlated if the Hurst exponent $H > 0.5$ (see Chapter 5), or anticorrelated if $H < 0.5$. In the uncorrelated percolation, the network's bonds are removed randomly. To preserve the correlations between the permeabilities, however, we remove those bonds that have been assigned the *smallest* permeabilities. The idea is that in a porous medium with a broad distribution of the permeabilities, a finite fraction of the medium's zones have very small permeabilities and, therefore, their contribution to the overall flow and transport properties of the medium would be negligible. Note that since for $H > 0.5$, the correlations are positive, most bonds with large or small permeabilities are clustered together. As a result, removal of the low-permeability bonds does *not* generate a tortuous cluster in the system. Moreover, precisely for the same reason, the percolation cluster generated by such a model does not have many dead-end bonds and is close to its backbone. This assertion is confirmed by the numerical results described below. On the other hand, if we consider the percolation cluster for $H < 0.5$, it is more tortuous and chaotic because the permeabilities are negatively correlated.

Several properties of the percolation model with long-range correlations were investigated using large-scale simulations and finite-size scaling (see Chapter 3). In particular, p_c , ν (the correlation length exponent), $\hat{\epsilon} = \epsilon/\nu$, $\hat{\mu} = \mu_p/\nu$, and the fractal dimensions D_f and D_{bb} of the sample-spanning cluster and its backbone were calculated. Here, ϵ and μ_p are the critical exponents associated with the power-law behavior of the permeability and conductivity near the percolation threshold (see Chapter 3). The results indicated that for $1/2 < H < 1$ – the range of interest here – the percolation threshold p_c decreases with increasing H , whereas the reverse is true for $0 < H < 1/2$. Moreover, ν and D_f essentially retain their value for random percolation, except when $H \simeq 1$, where $D_f \rightarrow 2$ (in 2D). It was also

found that for $H > 1/2$, D_{bb} increases with H and that $D_{bb} \rightarrow 2$ as $H \rightarrow 1$, that is, the cluster becomes compact. Moreover, $D_f \simeq D_{bb}$, confirming the assertion that for $H > 1/2$, the cluster and its backbone are similar. Table 11.1 presents the results for $1/2 < H < 1$, which, unlike random percolation for which the critical exponents are universal, indicates a smooth dependence of the exponents on H .

The correlated percolation model provides a rational explanation for the field-scale data for dispersion in the FS porous media. As emphasized earlier, if the permeabilities are distributed according to a FBM, then the pore space must contain zones of very low permeabilities, the elimination of which gives rise to a correlated percolation cluster with a backbone very close to the sample-spanning cluster. In fact, the backbone becomes identical with the cluster for $H \rightarrow 1$ (see Table 11.1). Therefore, since the fraction of the dead-end pores or stagnant regions in the system is very small, molecular diffusion that transfers the solute into and out of such regions plays no significant role. This means that dispersion is dominated by the stochastic velocity field imposed on the medium by the permeability distribution and, consistent with the field data, D_L depends on the average flow velocity V as

$$D_L \sim \xi V, \quad (11.200)$$

where ξ is some appropriate length scale. Under such conditions, the role of diffusion is to transfer the solute out of the slow boundary layer zones near the pore surfaces, and its effect only appears as a logarithmic correction to Eq. (11.200) (see Eq. (11.65)). Note that had we not removed the low permeability zones, diffusion into and out of such zones would have been important, and Eq. (11.50) would have implied that $D_L \sim V^2$, contradicting the field data.

Because the flow only takes place in the backbone of a percolation cluster, we should consider dispersion in the backbone of a correlated percolation cluster. Since the permeabilities are infinitely correlated, their correlation length is larger than any other relevant length scale of the system and, therefore, the only relevant length scale of the system is its linear size L , implying that the system is a backbone fractal for *any* L . Thus, Eqs. (11.90) and (11.93) should be used, implying that the exponent ν of Eq. (11.198) is given by $\nu = 1/(1 + \theta_B)$, except that when calculating θ_B , we must use the numerical values of the relevant exponents given in Table 11.1.

Table 11.1 Values of the critical exponents for 2D percolation with long-range correlation as a function of the Hurst exponent H .

H	$\hat{\epsilon}$	$\hat{\mu}$	D_{bb}
0.50	0.98	0.98	1.64
0.60	0.91	0.95	1.82
0.75	0.86	0.80	1.85
0.90	0.82	0.50	1.89
0.98	0.76	0.32	1.96

Another aspect of the model is that it is the 2D correlated percolation that is relevant to the field-scale data since such data are obtained at large distances from the source (up to several tens of kilometers), whereas the thickness of such porous media is at most a few hundred meters and, therefore, such porous media are long and thin and, thus, are essentially two-dimensional. Since the analysis of various field-scale permeability data by Hewett (1986) and others have indicated that $H > 1/2$ (and mostly $0.7 < H < 0.9$; see Chapter 5), from Table 11.1, we obtain a *nonuniversal* $\nu = 1/(1 + \theta_B) \simeq 0.53\text{--}0.62$ for this range, consistent with the field data described earlier. Note that if we do not remove the low permeability zones, we would have to use Eq. (11.101), which would yield estimates of ν that are not in the range of the field data. For example, we would obtain $\nu(H = 0.6) \simeq 2$, completely inconsistent with the field data.

Thus, percolation with long-range correlation is relevant to flow phenomena in field-scale porous media, for example, oil reservoirs and groundwater aquifers, as it provides a sound explanation for the field-scale data on the dispersion coefficients and dispersivities.

11.16

Dispersion in Unconsolidated Porous Media

Dispersion in flow through consolidated porous media is similar to that in unconsolidated ones and, therefore, all theories of dispersion described earlier are equally applicable to what we discuss in this section. Over the years, there have been many experimental studies of dispersion in flow through a packed bed. Some of the oldest works that the author is aware of are those of Ebach and White (1958), Carberry and Bretton (1958), Blackwell *et al.* (1959), Grane and Gardner (1961), Blackwell (1962), Harleman and Rumer (1963), Pfannkuch (1963), Edwards and Richardson (1968), Hassinger and von Rosenberg (1968), and Gunn and Pryce (1969). These works reported extensive experimental data for the longitudinal and transverse dispersion coefficients in a variety of packed beds using a wide variety of fluids, and over several orders of magnitude in the Péclet number Pe . Their results for both dispersion coefficients are completely similar to those shown in Figures 11.5 and 11.6.

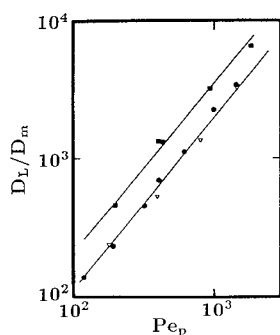


Figure 11.13 The effect of particle size distribution on the longitudinal dispersion coefficient D_L in packed beds. D_m is the molecular diffusivity and Pe_p is the particle Péclet number (after Han *et al.*, 1985).

As pointed out earlier, a crucial question about dispersion in flow through any kind of porous media is the condition(s) under which the dispersion coefficients become independent of time. This issue was studied by Han *et al.* (1985) for dispersion in flow through a packed bed. If a *particle Péclet number* is defined by

$$\text{Pe}_p = \frac{V_s d_p}{D_m} \frac{\phi}{1 - \phi}, \quad (11.201)$$

where D_m is the molecular diffusivity, then Han *et al.* (1985) showed that constant dispersion coefficients are obtained when a dimensionless time t_d

$$t_d = \frac{1}{\text{Pe}_p} \frac{L}{d_p} \frac{1 - \phi_p}{\phi_p} \gg 0.3, \quad (11.202)$$

where L is the length of the bed, and ϕ_p is the particles' volume fraction. Han *et al.* (1985) also studied the effect of a particle size distribution on the dispersion coefficients. Figure 11.13 presents their results for D_L for three types of beds. One was a uniform bed in which all the spherical particles had the same size, while the other two contained a range of particle sizes, but with the same average particle size as the uniform porous medium. The porosities of all the three porous media are also the same. As Figure 11.13 indicates, a broader particle size distribution gives rise to larger values of D_L , which is expected because a broader distribution generates more tortuous transport paths, and thus larger D_L . On the other hand, the results of Han *et al.* (1985) indicate no appreciable effect of the particle size distribution on the transverse dispersion coefficient.

Unlike the problem of flow and conduction through packed beds described in Chapter 9, theoretical investigation of dispersion through packed beds has not received a lot of attention. This is partly because theoretical studies of dispersion in *random* packings of particles is too complex and, as far as dispersion phenomena are concerned, regular or periodic arrays of particles do not provide realistic models of unconsolidated porous media. Such porous media do not have the type of tortuous flow paths that random packings have and, as emphasized earlier, dispersion is sensitive to the microstructure of porous medium. A spatially-periodic array of particles does not allow any disorder or heterogeneities.

Brenner (1980), Brenner and Adler (1982), Carbonell and Whitaker (1983), Eidsath *et al.* (1983), Koch *et al.* (1989), and Salles *et al.* (1993) studied dispersion in spatially-periodic systems. For example, Koch *et al.* (1989) showed that at high Péclet numbers, the mechanical dispersion that is caused by a stochastic velocity field is absent because flow in a spatially-periodic medium is completely deterministic, and at high values of Pe , molecular diffusion that can generate some microscopic stochasticity in the solute paths is not important. Thus, both D_L and D_T were shown to depend *quadratically* on the Péclet number, in contradiction with the experimental data for dispersion in *disordered* porous media – including *random* packings of particles – that indicate a much weaker dependence of the coefficients on the Péclet number.

Eidsath *et al.* (1983) carried out numerical simulations of dispersion in a square array of parallel cylinders in which the flow was parallel to the axis of the cylinders.

Using a finite-element method, they first solved for the flow field, and then solved the convective-diffusion equation. From their numerical simulations, Eidsath *et al.* found that $D_L \sim Pe^{1.7}$, which is closer to the experimental data described earlier in this chapter. From a theoretical view, however, their result cannot be explained because at high Pe , D_L must depend on Pe quadratically. Moreover, at very low Pe (i.e., when the flow field essentially has vanished), the results should be compatible with the conduction calculations described in Chapter 9. However, the results reported by Eidsath *et al.* were also not compatible with the limit of very low Pe . It is not known whether these discrepancies are due to numerical inaccuracies, to not having achieved the high- Pe limit, or reflect some other form of error.

11.17

Dispersion in Stratified Porous Media

The last topic to be briefly discussed is dispersion in stratified porous media. Field-scale porous media are usually stratified, consisting of many layers. The structural, and the flow and transport properties may vary greatly from stratum to stratum. For this reason, dispersion in stratified porous media has always been of interest. The earliest studies on transport in stratified porous media appear to be those of Koonce and Blackwell (1965) and Goddin *et al.* (1966), who studied the displacement of oil by water or a solvent in a stratified porous medium. Such works will be described in Chapter 14.

Marle *et al.* (1967) and Güven *et al.* (1984, 1985) applied the Taylor–Aris dispersion theory described above to a system of N strata that communicate with one another. Marle *et al.* (1967) obtained an integral expression for the longitudinal dispersion coefficient involving the porosity, fluid velocity, and the *local* transverse dispersion coefficient that were all functions of the distance perpendicular to the strata. Lake and Hirasaki (1981) and van den Broeck and Mazo (1983, 1984) also considered the Taylor–Aris dispersion in a stratified medium. In particular, Van den Broeck and Mazo derived several interesting results, including the FPTD and the longitudinal dispersion coefficient. Gelhar *et al.* (1979)’s model described earlier is, in fact, a method for studying dispersion in 2D stratified porous media since their equations represent averages over the vertical distance z , and the permeability field is assumed to depend on the distance perpendicular to the strata. Plumb and Whitaker (1988a,b) used their large-scale volume-averaging method mentioned earlier to study dispersion in stratified porous media.

In a seminal paper, Matheron and de Marsily (1980) studied dispersion analytically in a 2D stratified porous medium using analytical methods and asymptotic expansions. The direction of the flow velocity was assumed to be parallel to the bedding and constant for a given stratum. It was further assumed that the component of the velocity along the direction of macroscopic flow field is a weakly-stationary stochastic process. The permeability was assumed to be an isotropic stochastic process and the medium was of infinite extent in *both* directions. Matheron and de Marsily (1980) showed that under such conditions, dispersion is *never* Gaussian.

The reason is that because the porous medium is infinitely large and heterogeneous, a traveling solute particle always samples new regions and strata with new heterogeneities. As a result, a Gaussian dispersion regime can never be reached. Matheron and de Marsily also showed that if dispersion is to be Gaussian, then the integral of the covariance of the velocity (or permeability) must be zero which is, however, almost never satisfied for most realistic situations. On the other hand, if the macroscopic flow is *not* strictly parallel to the stratification (i.e., a small but finite perpendicular flow component is added), then dispersion will asymptotically be Gaussian if the integral is finite.

Bouchaud, J.P. *et al.* (1990) extended Matheron and de Marsily's work by studying a random walk in a 2D stratified medium containing a random velocity field. If the velocities in the x -direction – the macroscopic direction of the flow – are a function of the vertical distance, then Bouchaud *et al.* showed that

$$\langle \Delta x^2 \rangle \sim t^{\frac{3}{2}}, \quad (11.203)$$

that is, dispersion is superdiffusive, as $D_L \sim \langle \Delta x^2 \rangle / t \sim t^{1/2}$ and grows with the time. They also showed that there are large sample-to-sample fluctuations. The probability density $P(x, t)$, when averaged over various environments (realizations of the medium), was found to be non-Gaussian. It is approximately given by

$$\langle P(x, t) \rangle \sim t^{-\frac{3}{4}} f\left(\frac{x}{t^{\frac{3}{4}}}\right), \quad (11.204)$$

where $f(u)$ is a scaling function with the properties that $f(u) \sim \exp(-u^\delta)$ for $u \gg 1$, with $\delta = 4/3$. These results once again demonstrate the non-Gaussian nature of dispersion in the FS and stratified heterogeneous porous media, and the inadequacy of the CD equation for describing it.

12

Single-Phase Flow and Transport in Fractures and Fractured Porous Media

Introduction

In the last three chapters, we described and analyzed single-phase flow and transport processes in heterogeneous porous media. The porous media considered were, however, characterized by a *single* family of transport paths, namely, those that were made exclusively of the pores. Transport in such porous media is, therefore, characterized by a single transport equation. In a great many cases, the description of a porous medium by a single family of transport paths or a single spatial distribution of porosity attributed to the family is inadequate and, hence, the transport processes *cannot* be described by a single transport equation. As already discussed in Chapters 6 and 8, most natural rock masses consist of interconnected and intertwined networks of fractures and pores, implying the existence of *two* distinct distributions of porosities. One porosity distribution is contributed by the fractures, while the pores contribute the second distribution. In some cases, for example, carbonate rock, one may need *three* degrees or distributions of the porosity for characterizing the rock. Although most of the porosity is contributed by the pores, the fractures provide the most effective flow and transport paths. Moreover, transport processes in the pores and fractures can be quite different. For example, if the fracture network at large length scales is sample-spanning, it may be thought of as the backbone (flow-carrying part) of the porous medium in which flow and transport occur, while the porous matrix may act as a capacitor that is charged by the exchange with the adjacent fractures.

In addition to fractures and fractured porous media that are the focus of this chapter, there are several other systems of scientific and industrial importance in which transport takes place through two or more distinct families of transport paths. An example is provided by porous catalysts that usually contain large pores – the *macropores* – and very small pores – the *micropores*. The existence of the two distinct types of pores gives rise to considerable complications in modeling transport in porous catalysts. For example, transport in the micropores is hindered (restricted) in comparison with that in the macropores because the size of the transporting molecules is often comparable with those of the micropores. Moreover, whereas it is easy for the molecules to enter the macropores from the micropores, the reverse

is not always true, hence implying that the rates of exchange between the two types of pores are not necessarily equal.

A second example is coalbed methane reservoirs, an increasingly important example of a porous medium with distinct families of transport paths. Such reservoirs consist of large fractures and very small pores. It is generally believed that the production of methane gas, originally adsorbed on the coalbed matrix, occurs by desorption from the matrix and its subsequent diffusion towards the fractures. The pores are, however, so small that they do not allow the influx of water from the fractures. Their sizes are also comparable with the size of the methane molecules. Thus, molecular transport in the fractures of coalbed methane reservoirs is very different from that in the pores.

In general, if there are N distinct families of transport paths, flow and transport in the porous system must be modeled by N coupled transport equations, with the coupling accounting for the *exchange* between the various families of transport paths. In this chapter, we study flow and dispersion in fractures and fractured porous media, which correspond to the $N = 2$ case. However, the case of fractured porous media with $N = 3$ is also briefly described.

12.1

Experimental Aspects of Flow in a Fracture

In many cases, the permeability of fractures has been measured as a function of the confining pressure. Thus, one needs a reasonable model of deformation of a fracture, so as to relate the permeability to the mean aperture b_m . Another important issue is the relation between the effective permeability of a fracture and its mean aperture. The earlier data and their analysis led, however, to contradictory conclusions. Lomize (1951) suggested the empirical relation,

$$K_e = \frac{1}{12} b_m^3 \left[1 + 6 \left(\frac{a}{b_m} \right)^{\frac{3}{2}} \right]^{-1}, \quad (12.1)$$

with a being the asperities' height. A considerable amount of older data for fracture with smooth or rough, but nonconducting, surfaces were analyzed by Witherspoon *et al.* (1980), while Kranz *et al.* (1979) and Witherspoon *et al.* (1980) also analyzed data for fractures with conducting surfaces. The former group studied the effect of large confining pressure on the permeability of Barre granite, while Witherspoon *et al.* (1980) studied flow in fractures that are generated in rock by a Brazilian test (see also Barton *et al.*, 1985). They concluded that for all the cases studied,

$$K_e = \frac{b_m^3}{12\mathcal{R}}, \quad (12.2)$$

with $1.0 < \mathcal{R} < 1.65$ accounting for the effect of the surface roughness. A similar conclusion was reached by Plouraboué (1996), who measured the permeability of granite fracture with the two surfaces being at varying distances.

The conclusion that $K_e \sim b_m^3$ was, however, disputed by Gale (1982) and Raven and Gale (1985), who measured the permeability of natural and man-made fractures in granite and concluded that it was sensitive to the internal structure of the fractures as well as their length. Moreover, their data deviated strongly from the cubic law, Eq. (12.2). The same conclusion was reached by Durham and Bonner (1994), who measured the surface topography and permeability of Westerly granite. Their data indicated that for small mechanical apertures the permeability deviates strongly from the cubic law. Only when the mechanical aperture was large (on the order of 1 mm), did the cubic law seem to be satisfied. On the other hand, the data of Schrauf and Evans (1986) could very well be fitted with Eq. (12.1). Olsson and Brown (1993) reported that for a fracture undergoing compression and shear, the effective permeability varies with the third power of the *mechanical* aperture, which is the measured aperture when the surfaces come into contact.

Gentier (1986) carried out an extensive study of the flow properties of granite fractures under a normal load as well as the mean aperture and the fraction A_c of the contact area. If a local hydraulic aperture b_h is defined by $b_h = (12K)^{1/3}$ (K is the local permeability), Gentier found that for one sample and during the first and fourth loading cycles, $b_h \sim 0.55b_m$, almost independently of A_c . However, her results for a second sample indicated that b_h/b_m decreased as A_c increased during both cycles. Flow properties of six real fractures were studied by Hakami and Barton (1990), who also measured the tortuosity τ , defined as the ratio of the length of a streamline in the fracture and the fracture's length. The tortuosity was reported to be low, about 1–1.34, which perhaps was due to the fractures not having any contact area. The permeability data of Hakami and Barton were then reanalyzed by Zimmerman and Bodvarsson (1996) who showed that they can be fitted to within 20% by

$$K_e = \frac{1}{12} b_m^3 \left(1 - \frac{3}{2} \frac{\sigma_b^2}{b_m^2} \right), \quad (12.3)$$

where σ_b^2 is the variance of the aperture distribution.

However, as described in Chapter 6, there is ample experimental evidence (see, for example, Brown and Scholz, 1985; Brown *et al.*, 1986; Brown, 1987a,b; Power *et al.*, 1987; Poon *et al.*, 1992; Schmittbuhl *et al.*, 1993a; Cox and Wang, 1993; Odling, 1994; Boffa *et al.*, 1998; Ponson *et al.*, 2007) that the internal surface of natural fractures in rock masses – and even induced fracture surface of heterogeneous materials (Sahimi, 2003b) – is very rough, with the roughness profile following self-affine fractal statistics described in Chapters 5 and 6.

More precisely, consider the internal surface of a fracture with a single-valued function $h(x, y)$ representing the surface height, with the coordinates (x, y) being in the fracture's mean plane. Assume that the surface does not have any overhangs; that is, the surface is *continuously* rough. As described in Chapters 5 and 6, self-affinity of a rough surface implies that it exhibits scale invariance under rescaling with *direction-dependent* rescaling factors, $x \rightarrow b_x x$, $y \rightarrow b_y y$ and $h \rightarrow b_h h$. We assume isotropy in the fracture's mean plane, which is typically the case, and write

$b_x = b_y = b$ and $b_h = b^H$. Thus,

$$h(x, y) = b^{-H} h(bx, by), \quad (12.4)$$

where H is the roughness or Hurst exponent defined in Chapters 5 and 6. For example, Schmittbuhl *et al.* (1993a) measured the height of a granitic fault surface as a function of the position along one-dimensional (1D) profiles, and showed that the profiles exhibit self-affinity or anisotropic scale invariance (see Chapter 6).

Earlier studies (for reviews see, for example, Sahimi, 1998, 2003b) indicated that the fracture surface of *disordered materials* is characterized by a more or less universal value of the roughness exponent, $H \simeq 0.75\text{--}0.8$, with very weak dependence on the orientation considered. In fact, Bouchaud, E. *et al.* (1990) even conjectured that $H \simeq 0.8$ represents a universal value. However, more recent and careful studies indicate nonuniversality of H . It has been suggested that the roughness exponent may take on two distinct values, depending on the type of the fractured materials. One, $H \simeq 0.8$, is for such materials as glass, cement, granite, and tuff (Brown *et al.*, 1986; Måløy *et al.*, 1992; Poon *et al.*, 1992; Schmittbuhl *et al.*, 1993a; Glover *et al.*, 1998; Auradou *et al.*, 2005; Matsuki *et al.*, 2006), and another one, $H \simeq 0.5$, for sandstones (Boffa *et al.*, 1998; Ponson *et al.*, 2007), calcite (Gouze *et al.*, 2003) and sintered glass beads (Ponson *et al.*, 2006). However, the estimate, $H \simeq 0.75\text{--}0.8$ was also reported for sandstones (Nasseri *et al.*, 2006).

As for the internal surface of the fracture of rock, the estimate, $H \simeq 0.85$, was reported by Schmittbuhl *et al.* (1993a) for granitic faults. Vickers *et al.* (1992), Cox and Wang (1993), Johns *et al.* (1993), and Odling (1994), however, analyzed extensive data for a variety of rock joints and reported nonuniversal values of the roughness exponent H in the range $0 < H \leq 0.85$. There is also considerable indirect experimental evidence for roughness of the internal surface of fractures. Significant experimental deviations from the cubic law for the volume flow rate $Q \propto b^3$ have been observed and attributed (Tsang and Witherspoon, 1981; Hakami and Larsson, 1996) to the roughness of the internal surface of fracture. A detailed discussion of this important point was given by Konzuk and Kueper (2004).

12.2

Flow in a Single Fracture

In the early work on modeling of flow through fractures (see, for example, Snow, 1969), the fractures were typically represented by channels between two parallel smooth plates of length L . The problem of fluid flow through a fracture network is then reduced to one in a network of such channels, which is equivalent to the pore network models that we described in Chapters 10 and 11 for unfractured porous media. The same type of model may also be used for studying transport through fractures, for example, electrical conduction in fractures that are saturated by an electrically-conducting fluid.

It should be clear that with the internal surface of fractures being rough, the task of simulation of fluid flow and transport in fractures with a realistic model for the

roughness of their internal surface is complex (Brown, 1995). Since *exact* analytical solutions of the problems of fluid flow and transport (such as dispersion) in a single fracture with rough internal surface are not available, numerical simulations must be utilized. Such numerical simulations are mostly of two types:

1. In one approach, the simulations are based on discretizing the governing equations by a finite-difference (FD) or finite-element (FE) method and solving the resulting set of equations. However, if the effect of surface roughness of the fracture is to be taken into account, the FD or FE grid must be highly resolved near the surface, which would then require very intensive computations. One may also use the boundary-element (BE) method, but the necessary computations will still be very intensive. We should keep in mind that simulating fluid flow in a single fracture is only the prelude to simulating the same in a 2D or 3D network of fractures with rough internal surfaces. Therefore, while the computations with the BE and FE methods for a *single fracture* may be quite manageable, they will not be so for a 3D fracture network.
2. As a more efficient alternative, one may use a lattice-Boltzmann (LB) method (Rothman and Zaleski, 1997), already described in Chapters 9 and 11 for flow and transport in porous media. The LB method is ideally suited for simulation of fluid flow in porous media and fractures that have very irregular solid boundaries. Gutfraind and Hansen (1995) and Zhang *et al.* (1996) were the first to use a lattice-gas method (see Chapter 9) to study fluid flow in a single fracture with self-affine internal surfaces. The LB method has also been utilized extensively, and will be described shortly.

In addition to the flow properties, the electrical conductivity of fractures saturated by, for example, brine are also of great interest since resistivity measurements are a standard tool for estimating the porosity of rock. Thus, one main issue of interest to us in this chapter is how the fracture surface roughness affects its fluid permeability, electrical conductivity, and the dispersion coefficient. As emphasized in Chapter 4, however, there is no *exact* relation between the permeability and conductivity and, therefore, it is best to compare the effective apertures, that is, the hydraulic, the electrical, and the average geometrical apertures of a fracture in order to understand how its surface roughness affects its flow and transport properties.

There have been several approximate analytical solutions for the problem of fluid flow in a single fracture with rough surfaces. We first note that if the distribution of the *local* log-permeability $\gamma = \ln K$ is an even function, then

$$K_e = \exp(\langle \gamma \rangle) . \quad (12.5)$$

Equation (12.5) is applicable if the local aperture b and, hence, the local permeability K are log-normally distributed (Spector and Indelman, 1998). Calculating the

average $\langle \gamma \rangle$ in Eq. (12.5), one obtains $\langle K \rangle \exp(-\sigma_y^2/2) + \mathcal{O}(\sigma_y^6)$ and, therefore,

$$K_e = \begin{cases} \langle K \rangle (1 - \frac{1}{2} \sigma_y^2 + \dots) \\ \langle K \rangle \left(1 - \frac{\sigma_K^2}{2 \langle K \rangle^2} + \dots \right) \\ \frac{1}{12} \langle b \rangle^3 \left(1 - \frac{3 \sigma_b^2}{2 \langle b \rangle^2} + \dots \right) \\ \frac{1}{12} \langle b^3 \rangle \left(1 - \frac{9 \sigma_b^2}{2 \langle b \rangle^2} + \dots \right) \end{cases} . \quad (12.6)$$

The first equation is independent of any spatial correlation. Plouraboué (1996) computed higher-order terms in the last equation. Renshaw (1995) and Zimmerman and Bodvarsson (1996) showed that the range of applicability of Eq. (12.5) is broader than a log-normal permeability distribution.

We now describe and discuss the various analytical approximations and solutions to the problem of fluid flow in a fracture with rough surface, after which we will describe the results of computer simulations.

12.2.1

The Reynolds Approximation

The *local* flux q in a fracture is given by $q = w b^3 \Delta P / (12 \mu L)$, where w and L are, respectively, the width and length of the fracture, ΔP is the pressure drop along the fracture, and μ is the fluid viscosity. The continuity equation, $\nabla \cdot \mathbf{q} = 0$, then yields the Reynolds equation:

$$\nabla \cdot (b^3 \nabla P) = 0 . \quad (12.7)$$

Use of the Reynolds approximation yields the following for the hydraulic aperture b_h ,

$$b_h^3 = \langle b^{-3} \rangle^{-1} . \quad (12.8)$$

It is then a simple matter to calculate values of b_h from the microstructure of a fracture and its rough surface. This would still be an approximation since it is assumed that the flow is 1D. While one may expect approximation (12.8) to be relatively accurate when the upper and lower surfaces of a fracture are widely separated, it may lose its accuracy when the two surfaces are close. To derive Eq. (12.8), we use the fact that the hydraulic conductance of a short segment of the fracture is proportional to b^3 . The conductances are in series, that is, the *inverse* of the hydraulic conductance of the entire fracture is proportional to the sum of the inverses of the b^3 values.

Nicholl *et al.* (1999) tested the validity of the Reynolds approximation by comparing its predictions with detailed and high-resolution experimental data in an analog

fracture. They reported that the difference between the two sets of data varied between 22 and 47%, hence indicating that the 3D character of the flow field within the fracture may, in some cases, be important, and cannot be neglected.

12.2.2

Perturbation Expansion

Hasegawa and Izuchi (1983) studied fluid flow in a fracture modeled by a channel between a flat plane and a rough wall, with the roughness being periodic, and developed a perturbation expansion for the solution, where the perturbation parameter was $\epsilon = b_m/\omega \ll 1$, with b_m being the mean separation (aperture) between the two walls, and ω being the wavelength of the roughness. A similar approach was developed by Kitanidis and Dykaar (1997), while Pozrikidis (1987) solved the same problem by using the BE method. Hasegawa and Izuchi assumed that the flat plane is at $z^- = 0$ and the rough surface at $z^+ = b(x)$, where x is the direction perpendicular to the two walls. All the variables were first made dimensionless using $x' = x/\omega$, $y' = y/b_m$, $b' = b/b_m$ and $Q' = Q/(v_m b_m)$, where Q is the volume flow rate, and $v_m = \Delta P b_m^3/(12\mu L)$ is the mean flow velocity. The Reynolds number of the flow was defined by $Re = \rho v_m L/\mu$. The momentum equation was then recast in terms of the dimensionless variables and the stream function $\psi(x', y')$. The (dimensionless) volume flow rate was eventually obtained:

$$Q' = c_0 + c_2\epsilon^2 + \mathcal{O}(\epsilon^3), \quad (12.9)$$

where

$$c_0 = \langle b'^{-3} \rangle^{-1}, \quad c_2 = -\frac{3}{10}c_0^2 \left(1 + \frac{13}{8085}Re^2 c_0^2 \right) \left\langle \frac{db'}{dx} \right\rangle \frac{1}{b'^3}, \quad (12.10)$$

where $\langle \cdot \rangle$ denotes a spatial average over ω . The first term of Eq. (12.10) represents the Reynolds approximation, Eq. (12.8). Assuming that the roughness profile is given by $b(x) = b_m[1 + \delta \cos(2\pi x/\omega)]$ with $-1 < \delta < 1$, one obtains

$$Q = -\frac{\langle b'^{-3} \rangle^{-1} \Delta P}{12\mu L} \left[1 - \frac{6\pi^2(1-\delta^2)}{5(2+\delta^2)} \delta^4 \left(1 + \frac{13}{8085}Re^2 \right) \epsilon^2 \right] + \mathcal{O}(\epsilon^3). \quad (12.11)$$

The maximum of the correction term is $0.66\epsilon^2$, hence confirming that the Reynolds approximation for the pressure distribution, Eq. (12.8), is accurate; see also the discussions by Zimmerman and Bodvarsson (1996).

12.2.3

Effective-Medium Approximation

In Chapters 9 and 10, we described the essence of the effective-medium approximation (EMA). Walsh *et al.* (1997) proposed an EMA for the effective permeability

of a fracture given by

$$\int \frac{K_e - K}{K_e + K} f(K) dK = 0, \quad (12.12)$$

where $f(K)$ is the statistical distribution of the local permeability of a fracture. Observe that Eq. (12.12) is the 2D analogue of the EMA derived in Chapters 9 and 10. Reinterpreting what we learned in Chapters 9 and 10, Eq. (12.12) predicts that K_e vanishes for a fractional contact area $A_c = 1/2$, which appears to agree with some data. Actual fractures contain correlations, and positive correlations typically reduce the percolation threshold (see Chapter 3), whereas Eq. (12.12) cannot take into account the effect of the correlations. Therefore, at least some of the discrepancies between the predictions of Eq. (12.12) and some data may be due to the correlations' effect.

Drazer and Koplik (2002) analyzed the EMA further. They superposed a lattice on the space between the upper and lower surface of the fracture in which the distance between two neighboring points was a constant ξ_n . The bonds of the lattice were either parallel or perpendicular to the mean flow. The local volume flow rate q per unit width between two neighboring points is

$$q = -\frac{(h \cos \theta)^3}{12\mu} \frac{\Delta p}{\frac{\xi_n}{\cos \theta}} = -g \Delta p, \quad (12.13)$$

where θ is the angle of the block with the mean flow direction. If z is the height difference between the two neighboring points, then $\cos^2 \theta = \xi_n^2 / (\xi_n^2 + z^2)$ and, thus, $g = g_0 [\xi_n^2 / (\xi_n^2 + z^2)]^2$, where $g_0 = h^3 / (12\mu \xi_n)$. Equation (12.12) is then utilized for determining the conductance g and effective conductance g_e in terms of the distribution of the variable z . Drazer and Koplik (2002) then assumed that the distribution of the heights z is Gaussian. If $x = z / \sigma_z(\xi_n)$ (σ_z is the standard deviation), then Eq. (12.12) yields

$$\int \frac{1}{\sqrt{2\pi}} \exp\left(-\frac{x^2}{2}\right) \frac{g_e(1 + \epsilon^2 x^2)^2 - g_0}{g_e(1 + \epsilon^2 x^2)^2 + g_0} dx = 0. \quad (12.14)$$

Here, $\epsilon = \sigma_z(\xi_n) / \xi_n = (\xi_n / l)^{H-1}$, with H being the roughness (Hurst) exponent. Since $\sigma_z(\xi_n) \ll \xi_n$ (l is a microscopic characteristic length), one must have $\epsilon \ll 1$. Thus, a perturbation expansion similar to Eq. (12.9) may also be utilized for the effective conductance, $g_e = a + b\epsilon^2 + c\epsilon^4 + \dots$ that, when substituted into Eq. (12.14), yields

$$g_e = g_0(1 - 2\epsilon^2 + 5\epsilon^4 + \dots), \quad (12.15)$$

which should be accurate if the upper and lower surfaces are not too close.

12.2.4

Asymptotic Expression

Similar to the Reynolds approximation, we consider a fracture with a rough surface consisting of N segments, where each segment or channel is oriented at some angle θ_i with respect to the mean plane with an effective aperture, $b_i = \langle b \rangle \cos \theta_i$, and length $\xi_{||}^i = \xi_{||} / \cos \theta_i$ (Drazer and Koplik, 2000). An example is shown in Figure 12.1. The roughness profile is assumed to be described by a fractional Brownian motion with the Hurst exponent H . It is also assumed that the segment of length $\xi_{||}$ in the direction of mean flow, over which the channel is formed by the two fracture surface, is approximately straight (see Figure 12.1).

The hydraulic conductance g_h^i of segment i of the channel is given by

$$g_h^i = \frac{b_i^3 w}{12 \xi_{||}^i}, \quad (12.16)$$

and, therefore, the effective hydraulic conductance g_h^e of the fracture is given by

$$(g_h^e)^{-1} = \sum_{i=1}^N (g_h^i)^{-1} = \frac{12 \xi_{||}}{w \langle b \rangle^3} \sum_{i=1}^N \cos^{-4} \theta_i = \frac{12 L}{w \langle b \rangle^3} \langle \cos^{-4} \theta \rangle. \quad (12.17)$$

Since the roughness exponent $H < 1$, $\cos \theta$ is given by

$$\cos \theta = \frac{\xi_{||}}{\sqrt{\xi_{||}^2 + C_z^2(\xi_{||})}}, \quad (12.18)$$

where $C_z(\xi_{||})$ is a correlation function such that $C_z^2 = \zeta(l)(\xi_{||}/l)^{2H}$, with l being a microscopic length, and $\zeta(l)$ being a function that is of the order of the microscopic length squared. By substituting Eq. (12.17) into the relation for the effective

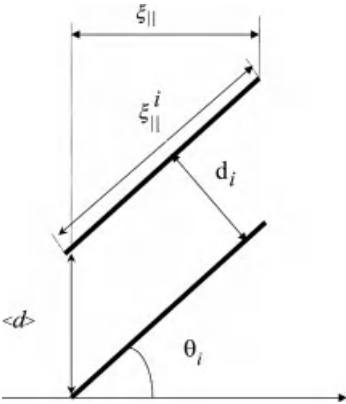


Figure 12.1 Approximation of the roughness of the fracture surface.

permeability, $K_e = (g_h^e L)/(\langle b \rangle w)$ and using Eq. (12.18), one obtains (Drazer and Koplik, 2000):

$$K_e = \frac{\langle b \rangle^2}{12} \left\{ 1 - 2 \left[\frac{\zeta(l)}{l^2} \right] \left(\frac{\langle b \rangle}{l} \right)^{\frac{2H-2}{H}} \right\}. \quad (12.19)$$

Thus, the hydraulic aperture b_h is given by

$$b_h = \langle b \rangle \left\{ 1 - 2 \left[\frac{\zeta(l)}{l^2} \right] \left(\frac{\langle b \rangle}{l} \right)^{\frac{2H-2}{H}} \right\}^{\frac{1}{3}}. \quad (12.20)$$

Drazer and Koplik (2000) stated that Eq. (12.20) is accurate for $H \approx 1$. However, Madadi *et al.* (2003) showed that if the results are expressed in terms of the various apertures described above, then, at least for a range of the values of the local apertures, Eq. (12.20) is accurate for all values of H . We will come back to this point shortly. Note that Eq. (12.19) is equivalent to Eq. (12.15) to the ϵ^2 order.

12.2.5

Effect of the Contact Areas

Both the perturbation expansion and the EMA breakdown if contact zones do exist in a fracture. However, whereas it would be very difficult to include the effect of the contact area in the perturbation expansion, the EMA does provide a solution for the problem by viewing the contact areas as having zero permeability and the rest following a permeability distribution, similar to percolation networks of resistors described in Chapter 10. Hence, Eq. (12.12) would predict that

$$A_c + (1 - A_c) \int \frac{K_e - K}{K_e + K} f(K) dK = 0. \quad (12.21)$$

If all the non-contacted areas have the same permeability K_0 , then Eq. (12.21) reduces to

$$K_e = K_0(1 - 2A_c). \quad (12.22)$$

Zimmerman and Bodvarsson (1996) combined Eqs. (12.6) and (12.22) to propose that

$$K_e = \frac{1}{12} \langle b \rangle^3 \left(1 - \frac{3\sigma_b^2}{2\langle b \rangle^2} \right) (1 - 2A_c). \quad (12.23)$$

Equation (12.23) was shown to predict the experimental data for K_e to within a factor of two for $0 < A_c < 0.35$.

12.2.6

Numerical Simulation

In addition to the analytical approximations, extensive computer simulation of flow in a single fracture has been carried out in the past. In what follows, we summarize the most important aspects of the results.

12.2.6.1 Mapping onto Equivalent Pore Networks

As described in Chapter 6, fracture aperture decreases under stress. It has also been observed that under stress, the fluid flow through a single fracture is channelized and usually along certain channels. Figure 12.2, taken from Billaux (1990), shows the image of the cross section of a single fracture with its contact and void areas under normal stress. Also shown is the same system after the dead-end channels and small loops of channels had been removed. The implication of Figure 12.2 is clear: *each* fracture should be mapped onto an equivalent network of channels, an example of which is also shown in Figure 12.2. Thus, flow calculations in a single channel can be mapped onto one in a network. The structure of the network depends, of course, on the morphology of the fracture, and in particular, on the distribution of its contact areas. Data for such characteristic properties must be obtained with careful (and often very tedious) measurements. Once the channel network is constructed, flow calculations in the network (in the fracture) are carried out, a subject already studied in Chapter 10.

Such an approach was developed by Tsang (1984), Moreno *et al.* (1988), Tsang *et al.* (1988), and Nolte *et al.* (1989). Figure 12.3, adopted from Moreno *et al.* (1988), shows the distribution of the fluid fluxes in an idealized 2D network model of a single fracture in which the apertures were distributed according to a log-normal distribution. The apertures were correlated with each other, with a spatial covariance that decayed exponentially. The thick lines show the channels through which most of the fluid flows, and are clearly indicative of the existence of channeling. Mapping flow through a single fracture onto flow through a network also implies that percolation theory is relevant even to flow through a single fracture, as stress decreases the void space of the fracture and creates flow channels. Moreover, as Figure 12.3 indicates, most of the fluid flows through a few channels and, thus, the connectivity of the channels is important to fluid flow through a single fracture. Indeed, as discussed earlier, the EMA does provide a means for evaluating the percolation threshold of a single fracture at which its effective permeability vanishes.

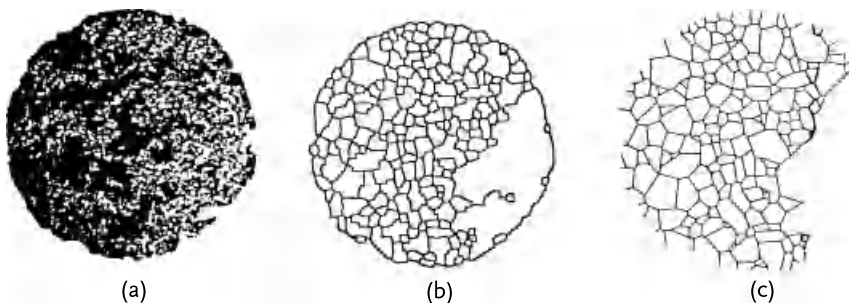


Figure 12.2 A binary image of a single fracture (a), its equivalent skeleton (b) in which the dead-end parts and small loops were removed, and its equivalent network (c). Dark areas represent the void space (after Billaux, 1990).

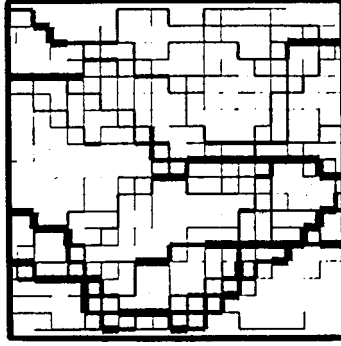


Figure 12.3 A 2D network representation of a single fracture and the distribution of flow rates in it. The thickness of the lines is proportional to the magnitude of the flow rates.

Observe that the effective network has the structure of a percolation backbone shown in Chapter 3 (after Moreno *et al.*, 1988).

12.2.6.2 Numerical Simulation of the Reynolds Equation

Instead of attempting to develop an approximate analytical solution of the Reynolds equation, Eq. (12.7), one may also obtain its complete solution through computer simulation. Patir and Cheng (1978) were probably the first to carry out numerical simulation of the Reynolds equation. They generated rough Gaussian random surfaces a distance b_m apart, in which the variance of the roughness height was σ_h^2 , and the correlations decreased linearly with the distance. From the numerical solution of Eq. (12.7), the effective permeability was computed. Their results could be fitted accurately to

$$K_e = \frac{1}{12} \langle b \rangle^3 \left[1 - 0.9 \exp \left(\frac{-0.56 b_m}{\sigma_h} \right) \right]. \quad (12.24)$$

More refined simulations were carried out by Brown (1987b, 1989) who used self-affine surfaces in order to generate the rough surface of a fracture. A pressure drop was applied to the fracture parallel to the surfaces, and the hydraulic aperture, $b_h = (12 K_e)^{1/3}$, was computed. Brown reported that if $b_h \geq \sigma_h$, then the roughness (Hurst) exponent H has a very weak effect on b_h . Moreover, $b_h / \langle b \rangle \rightarrow 1$ for large apertures, whereas $b_h / \langle b \rangle \rightarrow 1/2$ for $b_m = \sigma_h$.

12.2.6.3 Numerical Simulations with a Three-Dimensional Fracture

Extensive computer simulations of flow in a single fractures were carried out by Mourzenko *et al.* (1995). The model utilized was an infinite periodic fracture that consisted of identical unit cells of size $l \times l$ in the (x, y) plane. The internal surface of the fracture was represented by the heights, $z^\pm = h_0 + h^\pm(x, y)$, above the reference planar $z = 0$, where $h^\pm$ were random periodic functions with Gaussian

probability density,

$$\varphi(h^\pm) = \frac{1}{\sqrt{2\pi}\sigma_h} \exp \left[-\frac{(h^\pm)^2}{2\sigma_h^2} \right]. \quad (12.25)$$

Thus, the mean distance $\frac{1}{2}(h^+ + h^-)$, and the distance $b = h^+ - h^-$ are also Gaussian variables. If $b < 0$, then the two surfaces are in contact. The mean separation is then, $b_m = \langle b \rangle$. The correlation function used for the rough surfaces was

$$C_h(r) = \begin{cases} \sigma_h^2 \exp \left(-\frac{r^2}{\xi_s^2} \right), & \text{Gaussian fracture, } H = 1 \\ \sigma_h^2 \left[1 - \left(\frac{r}{l_c} \right)^{2H} \right], & \text{self-affine fracture, } H < 1 \end{cases} \quad (12.26)$$

where ξ_s is the correlation length, and l_c is a characteristic length scale. The space between the rough surfaces was then discretized by the method described in Chapter 8. Both the Stokes and Reynolds equations were solved numerically in the space between the surfaces. If the fractional contact area A_c was small, the flow patterns obtained by the two equations were similar. For comparison, the permeabilities $K_1 = \langle b \rangle^3/12$ and $K_2 = b_m^3/12$ for flow between flat plates were also computed, and the ratios $\langle K_e^{(i)} / K_1 \rangle$ were computed where $i = S$ (for Stokes' equation) and $i = R$ for the Reynolds equation.

The ratio $K_e^{(R)} / K_1$ was found to be insensitive to (1) the slope of the spectral density of the rough surface that defines the correlations (see Chapter 5); (2) l_c / σ_h , and (3) close to one for $b_m / \sigma_h > 6$ and all the values of σ_h . The same was not true for $K_e^{(S)} / K_1$; even for $b_m / \sigma_h = 10$, the ratio was still significantly less than one, and decreased with decreasing $1 / \sigma_h$. The flow rate derived from the Reynolds equation was always larger than that from the Stokes equation by a factor larger than two. The solutions from the two equations agreed better when the characteristic length l_c was large.

12.2.6.4 Lattice-Gas and Lattice-Boltzmann Simulations

Gutfraind and Hansen (1995) and Zhang *et al.* (1996) were the first to use a lattice-gas (LG) method to study fluid flow in a single fracture with self-affine internal surfaces. The lattice-Boltzmann (LB) method was also utilized by Drazer and Koplik (2000, 2002), Madadi *et al.* (2003), Madadi and Sahimi (2003), and Kim *et al.* (2003). Both methods were described in detail in Chapter 9. Thus, it suffices to say that, once a model of a fracture with rough internal surface is generated and a lattice structure is superimposed onto the space between the fracture's surface, the LB or LG method described in Chapter 9 is used to simulate the fluid flow and compute the effective permeability.

Consider the hydraulic aperture b_h , and let us compare the results obtained from the LB simulations with the predictions of Eq. (12.20) and the Reynolds approximation as well as the average aperture $\langle b \rangle$ (Madadi *et al.*, 2003). The results for the roughness exponent $H = 0.25$, presented in Figure 12.4, indicate that for

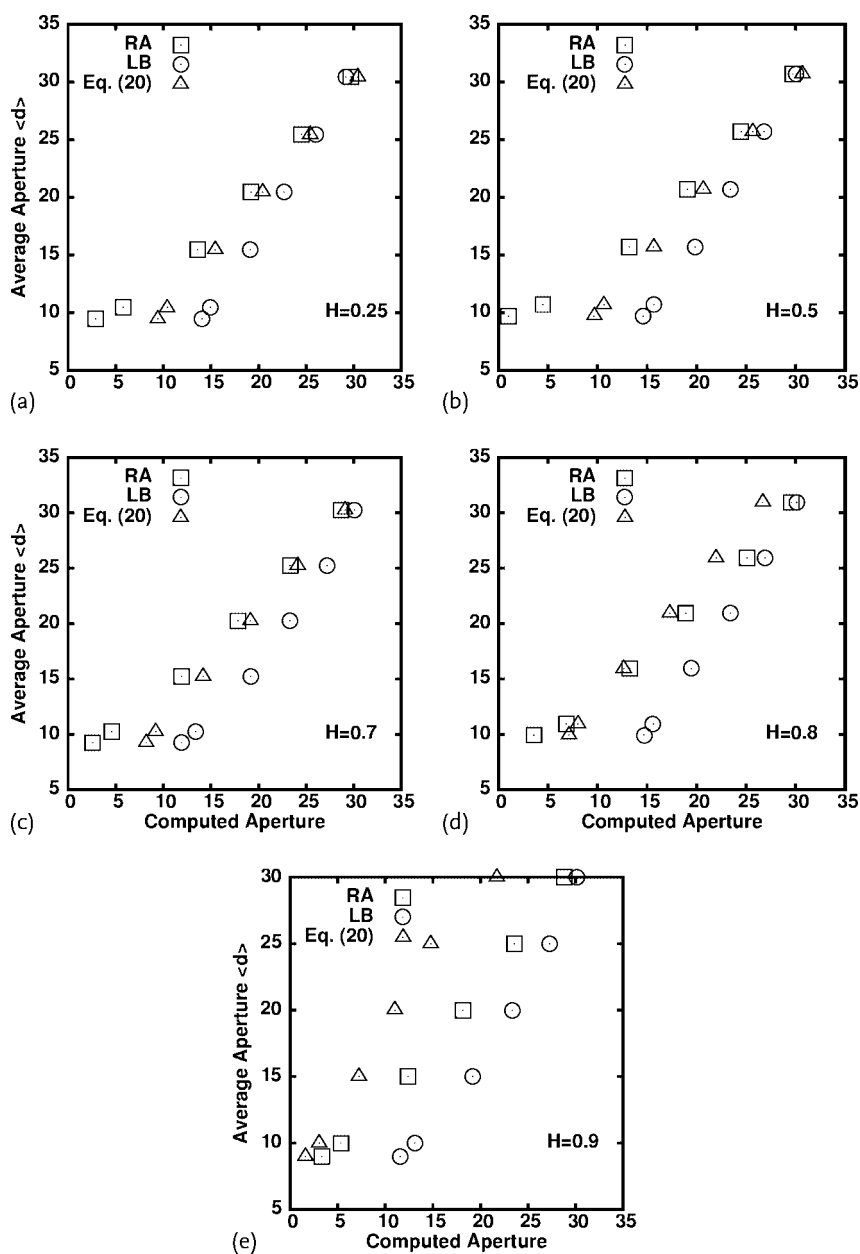


Figure 12.4 Comparison of the hydraulic apertures b_h , computed by the Reynolds approximation (RA) and the lattice-Boltzmann (LB) method, with the corresponding geo-

metrical apertures and the predictions of the asymptotic expression, Eq. (12.20). (a) $H = 0.25$; (b) $H = 0.5$; (c) $H = 0.7$; (d) $H = 0.8$; (e) $H = 0.9$ (after Madadi *et al.*, 2003).

wide fractures, the three methods yield very similar estimates, all of which are close to $\langle b \rangle$, whereas for narrow fractures, there are significant differences between the three estimates. In particular, the Reynolds approximation provides very poor estimates of b_h because the flow field is no longer 1D (see also below), whereas the estimates obtained from the LB simulations are larger than the predictions of Eq. (12.20) and $\langle b \rangle$. The same trends are seen for $H = 0.5$.

The trends are somewhat different for $H > 0.5$ (positive correlations). Consider, for example, the case of $H = 0.7$, the results for which are also shown in Figure 12.4. Once again, for wide fractures, the three methods yield estimates of b_h that are close to one another as well as to $\langle b \rangle$, with Eq. (12.20) yielding the largest estimate and the Reynolds approximation predicting the smallest values. For narrow fractures, the Reynolds approximation is again very poor. The same trends continue to hold for $H = 0.8$. For $H = 0.9$, there are significant differences between the various estimates, even when the fractures are wide. Thus, if we view the estimates provided by the LB method to be the most accurate and reliable values of the hydraulic apertures, then the results shown in Figure 12.4 indicate that the Reynolds approximation breaks down completely for even relatively narrow fractures.

12.3

Conduction in a Fracture

Similar to single-phase flow in a fracture, the problem of conduction in a fracture saturated by a conducting fluid (such as brine) has also been studied extensively. Bernabé (1982), Batzle and Simmons (1983), and Stesky (1986) reported the results of extensive experimental works on the conductivity of fractures. Stesky found that the fracture's effective permeability and conductivity are related:

$$K_e \propto g_e^3. \quad (12.27)$$

Earlier, Walsh and Brace (1984) had proposed that

$$K_e \propto g_e^\zeta, \quad (12.28)$$

with $1 \leq \zeta \leq 3$.

If the distribution of the log-conductivity, $\gamma = \log g$, is an even function, then Eqs. (12.5) and (12.6) are also applicable to the effective conductivity g_e . In particular,

$$\frac{g_e}{g_0} = \langle b \rangle \left(1 - \frac{1}{2} \sigma_Y^2 + \dots \right) = \langle b \rangle \left(1 - \frac{\sigma_b^2}{2\langle b \rangle} + \dots \right). \quad (12.29)$$

Note that the EMA may be used for computing the effective conductivity as well. In what follows, we describe the most important aspects of the problem and the results.

12.3.1

The Reynolds Approximation

We proceed in a manner similar to that for the fluid flow problem. The local current $\mathbf{I}$ in a fracture is given by $\mathbf{I} = -bg_0\nabla V$, where V is the potential and g_0 is the conductivity of the fluid. The continuity equation, $\nabla \cdot \mathbf{I} = 0$, then yields the Reynolds equation

$$\nabla \cdot (b\nabla V) = 0. \quad (12.30)$$

Thus, the Reynolds approximation yields the following result for the electrical aperture b_e of fractures:

$$b_e = \langle b^{-1} \rangle^{-1}. \quad (12.31)$$

Derivation of Eq. (12.31) is similar to that used for deriving Eq. (12.8). More generally though, $\langle b^{-1} \rangle \leq b_e \leq \langle b \rangle$.

12.3.2

Perturbation Expansion

Consider a 2D fracture with surfaces that are bounded by the lower and upper surfaces, $z^-(x, y) = \sigma_h Z^-(x, y)$ and $z^+(x, y) = b_m + \sigma_h Z^+(x, y)$, with $\epsilon = \sigma_h/b_m \ll 1$. Volik *et al.* (1997) solved Eq. (12.30) with $Z^- = 0$ and $Z^+(x, y) = \sqrt{2} \sin(\omega x)$, where $\omega = 2\pi/L$ with L being the fracture's length. They derived the following equation:

$$\frac{g_e}{g_0} = b_m [1 - \epsilon^2 \coth(\omega b_m) + \dots]. \quad (12.32)$$

If the roughness profile of the upper surface is random with a 1D spectral density $S_1(\omega_n)$,

$$S_1(\omega_n) = \frac{2}{L} \int_0^L R(x) \cos(\omega_n x) dx, \quad (12.33)$$

where $R(x)$ is the covariance (see Chapter 5), then

$$\frac{g_e}{g_0} = b_m \left[1 - \epsilon^2 b_m \sum_{n=1}^{\infty} \omega_n \coth(\omega_n b_m) + \dots \right], \quad (12.34)$$

which, in the long wavelength limit, $\omega b_m \rightarrow 0$ reduces to

$$\frac{g_e}{g_0} = b_m (1 - \epsilon^2 \dots) = b_m \left(1 - \frac{\sigma_h^2}{b_m^2} \right). \quad (12.35)$$

If the roughness profile of the lower surface is random and uncorrelated with the upper one, Eq. (12.34) is still applicable, except that the ϵ^2 term should be multiplied by a factor of two.

Mourzenko *et al.* (1999) generalized the results of Volik *et al.* (1997) to 3D fractures with $Z^- = 0$ and a random Z^+ with a given 2D spectral density, $S_2(\omega)$. Their result is given by

$$\frac{g_e}{g_0} = b_m \left[1 - \epsilon^2 \left(\frac{2\pi b_m}{L^2} \right) \sum_i \sum_j \left(\frac{\omega_n^2}{\omega} \right) S_2 \left(\frac{i}{L}, \frac{j}{L} \right) \coth(2\pi \omega b_m) + \dots \right], \quad (12.36)$$

where $\omega = |\omega|$. If S_2 decays rapidly with ω , then Eq. (12.36) is simplified to

$$\frac{g_e}{g_0} \approx b_m \left(1 - \frac{1}{2} \epsilon^2 \right). \quad (12.37)$$

12.3.3

Asymptotic Expression

An equation similar to Eq. (12.20) may be derived for the aperture b_e (Madadi *et al.*, 2003). The electrical conductance g_i of segment i of the channel with aperture b_i is given by

$$g_i = \frac{g_0 b_i w}{\xi_{||}^i}, \quad (12.38)$$

where the notation is the same as before. Then, noting that $N = L/\xi_{||} \gg 1$ (N is the number of the segments along a fracture), the effective conductance g_e of the fracture is given by

$$g_e^{-1} = \sum_{i=1}^N g_i^{-1} = \frac{\xi_{||}}{g_0 \langle b \rangle w} \sum_{i=1}^N \cos^{-2} \theta_i = \frac{L}{g_0 \langle b \rangle w} \langle \cos^{-2} \theta \rangle. \quad (12.39)$$

By substituting Eq. (12.18) in Eq. (12.39), one obtains

$$g_e^{-1} = \frac{L}{g_0 \langle b \rangle w} \left\{ 1 + \left[\frac{\zeta(l)}{l^2} \right] \left(\frac{\langle b \rangle}{l} \right)^{\frac{2H-2}{H}} \right\}. \quad (12.40)$$

Therefore, the effective electrical aperture b_e is given by

$$b_e = \langle b \rangle \left\{ 1 + \left[\frac{\zeta(l)}{l^2} \right] \left(\frac{\langle b \rangle}{l} \right)^{\frac{2H-2}{H}} \right\}. \quad (12.41)$$

12.3.4

Random Walk Simulation

Let us first mention that Volik *et al.* (1997) and Mourzenko *et al.* (1999, 2001) carried out extensive computer simulations of conduction in fractures with rough surfaces.

In particular, Mourzenko *et al.* simulated the case in which the roughness profile of the fracture surface is self-affine with the roughness (Hurst) exponent H .

A simpler and equally accurate method for computing the conductivity of a fracture is based on random walk (RW) simulation (van Sickle, 2002a; Madadi *et al.*, 2003; see also Chapters 9 and 10). The method exploits the isomorphism between the conduction equations and the diffusion equation for a collection of non-interacting random walkers in the presence of a driving force. The phase domains in a composite microstructure correspond to distinct populations of walkers, where the equilibrium walker density $\rho_w(\mathbf{r})$ of a population is given by the value of the transport coefficient $g(\mathbf{r})$ of the corresponding phase domain. The principle of detailed balance ensures that the population densities are maintained, and provides the following rule for the RW over a scalar field (or digitized, or discretized, microstructure): A walker at site (or pixel) i , in a d -dimensional space, attempts a move to a randomly selected adjacent site j during the time interval $(4d)^{-1}$ in 2D or $(6d)^{-1}$ in 3D. The move is successful with probability

$$W_{ij} = \frac{g_j}{g_i + g_j}, \quad (12.42)$$

where g_i and g_j are the conductivities of the phases at sites i and j , respectively. The effective conductivity g_e is related to the effective diffusivity D_e by

$$g_e = \langle g(\mathbf{r}) \rangle D_e, \quad (12.43)$$

where $\langle g(\mathbf{r}) \rangle$ is the volume average of the constituent transport coefficients, and D_e is given by the usual equation,

$$D_e = \frac{\langle R^2 \rangle}{2dt}, \quad (12.44)$$

where $\langle R^2 \rangle$ is the mean-square displacement of the walker over the time interval t , with the average taken over all the initial positions of the walkers as well as all the possible realizations of the system.

In the calculations of the electrical conductivity by the RW method (which effectively solves the Eq. (12.30) over the interior of the fracture), one specifies equipotential surfaces at the two ends. A square or simple-cubic lattice is placed over the d -dimensional fracture. A conductivity of one is attributed to sites completely contained within the void space of the fracture, whereas a conductivity of zero is assigned to the sites lying completely outside the fracture (in its walls). To those sites intersected by a fracture surface, a conductivity equal to the fraction of the site lying within the fracture is assigned. Therefore, if, for example, only a fraction 1/3 of a square site lays within the fracture, a conductivity 1/3 is assigned to that site. One could, of course, choose other reasonable rules.

Let us now compare the predictions of the asymptotic expression for b_e , Eq. (12.41), with the Reynolds approximation and the RW simulations (Madadi *et al.*, 2003). As Figure 12.5 indicates, for the roughness exponent $H = 0.25$

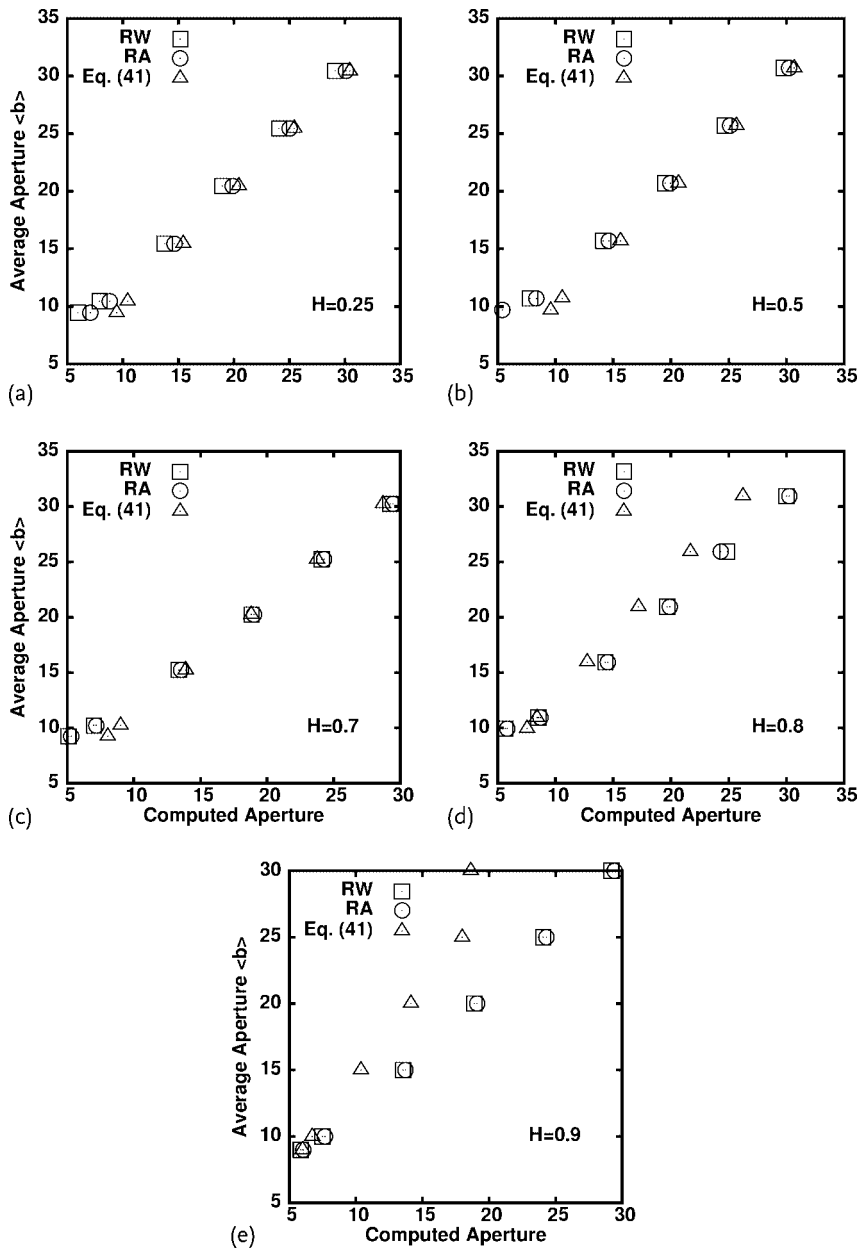


Figure 12.5 Comparison of the electrical apertures b_e computed by the RW method and the Reynolds approximation (RA), with the corresponding mean geometrical apertures and

the predictions of the asymptotic expression, Eq. (12.41). (a) $H = 0.25$; (b) $H = 0.5$; (c) $H = 0.7$; (d) $H = 0.8$; (e) $H = 0.9$ (after Madadi *et al.*, 2003).

and wide fracture (large $\langle b \rangle$), the RW method, the Reynolds approximation, and Eq. (12.41) all yield practically the same estimates of b_e . However, whereas for narrow fractures Eq. (12.41) predicts that $b_e \simeq \langle b \rangle$, the Reynolds approximation and the RW method provide estimates of b_e that are significantly lower than $\langle b \rangle$. In particular, the RW method, which is usually very accurate, provides estimates that are much lower than $\langle b \rangle$, indicating that using a simple average $\langle b \rangle$ for computing the effective conductivity of a fluid-saturated fracture results in gross error. The same trends are seen for $H = 0.5$.

For $H > 0.5$, however, the trends change qualitatively. Consider, for example, the results for $H = 0.7$, also presented in Figure 12.5. Once again, for wide fractures with large mean apertures $\langle b \rangle$, the predictions of the Reynolds approximation and Eq. (12.41) are very close to the estimates obtained by the RW method, and all the three estimates are also close to $\langle b \rangle$. However, as the fracture becomes narrower, significant differences between the three estimates emerge, except that, compared with the cases of $H \leq 0.5$, the prediction of Eq. (12.41) is somewhat closer to the RW estimate, and begins to be significantly different from $\langle b \rangle$. These trends also hold as the roughness exponent H increases and approaches one, that is, as the fracture's surface becomes completely smooth. In the case of $H = 0.9$, the Reynolds approximation and the RW method yield very similar estimates for the electrical aperture b_e , which are significantly different from the prediction of Eq. (12.41) if the fracture is wide. However, for narrow fractures, Eq. (12.41) predicts values of b_e that are very close to those obtained from the Reynolds approximation and the RW method, with the three estimates being much lower than the average aperture $\langle b \rangle$.

Several conclusions may be drawn from Figures 12.4 and 12.5:

1. Using a simple mean aperture for representing a fracture with rough internal surfaces will always result in gross errors unless, of course, the fractures are wide, in which case the surface roughness does not really matter.
2. For both the conduction and fluid flow problems, the Reynolds approximation is relatively accurate only when the fractures are either very wide or at least moderately so. For narrower fractures, the streamlines (in the flow problem) and the equipotential lines (in the conduction problem) are too curved and complex, hence breaking down the assumption of a quasi-1D flow or transport problem used in the Reynolds approximation. For such fractures, the Reynolds approximation breaks down completely.
3. For H close to unity, the asymptotic expression (12.41) for the effective electrical aperture is very accurate for narrow and relatively wide fractures, whereas for smaller values of H , Eq. (12.41) provides accurate estimates of the effective electrical aperture for wide and moderately narrow fractures.
4. The same type of trends are generally predicted by Eq. (12.20) for the effective hydraulic apertures. However, for $H \approx 1$, even for moderately wide fractures, the predictions of Eq. (12.20) are not in agreement with the numerical estimate of b_h obtained by the LB method.

5. The effective electrical and hydraulic apertures of a fracture are very different, hence indicating again why there is no general relation between the effective permeability and conductivity of a fluid-saturated fractured or unfractured porous medium, or even a single fractures with rough, self-affine surfaces.
6. The LB and RW results indicate that, in all cases, the effective electrical aperture b_e is less than b_h , the effective hydraulic aperture. This is what one may expect for real 3D fractures since, relatively speaking, a larger electric flux flows through the narrow parts of the fracture than the fluid flux and, therefore, the electric flux perceives a narrower fracture than the fluid flux does. Indeed, the computations by van Sichen (2002a), who used the Reynolds approximation for both the electrical conduction and fluid flow problems, indicated that in 3D, $b_e < b_h$. For the 2D fractures for which the above comparisons were made, the result that $b_e < b_h$ may reflect greater tortuosity of the electric flux lines than the fluid streamlines.

12.4

Dispersion in a Fracture

Dispersion in flow through fractures is important to oil recovery processes, groundwater flow, and the spread of contaminants in groundwater and, hence, has received considerable attention from researchers in both fields. However, as discussed in Chapter 11, dispersion is a far more complex phenomenon than the problem of flow in a porous medium or in fractures. Chapter 11 already described the important elements of dispersion in flow through porous media, and the various computational and theoretical approaches to it. The same fundamental concepts and modeling approaches can be used to study and understand dispersion in a fracture, with the added complexity of the effect of the fracture's rough walls.

12.4.1

Experimental Aspects

Several groups have reported experimental data for tracer experiments in a single fracture measured both in laboratory and under field conditions. They include the early works of Grisak and Pickens (1980), Neretnieks (1980), Tang *et al.* (1981), Noorishad and Mehra (1982), Novakowski *et al.* (1985), Raven *et al.* (1988), and Lowell (1989). These studies were concerned with the effect of the matrix on transport through a single fracture, and how experimental data should be interpreted. Some of the works also involved analytical analysis as well as numerical simulations. They are useful for modeling transport in a network of fractures because simulation of flow and transport in a fracture network requires the solution of the same problems at the level of a single fracture.

Neretnieks *et al.* (1982) studied migration of radionuclide in a natural fracture in a granite core that was parallel to the axis of the cylindrical core. Both sorbing and nonsorbing tracers were used. The breakthrough curves for the nonsorbing tracers indicated that channeling may occur, while those for the sorbing tracers indicated that diffusion into the porous matrix is significant.

Grisak *et al.* (1980) injected a tracer solution containing Cl and Ca ions into a large cylindrical sample of fracture obtained in a fresh excavation. The sample contained two orthogonal sets of fractures parallel to the long axis of the cylinder. The breakthrough curves for both ions indicated significant retardation of the ions relative to the average water velocity in the fracture. Grisak *et al.* (1980) attributed the retardation to the effect of diffusion of the solutes from the fracture into the porous matrix of the till.

Moreno *et al.* (1985) carried out dispersion experiments in a sample natural fracture. They used two granitic drill cores taken from Stripa mine, each containing a natural fracture running parallel to the axis. Both sorbing and non-sorbing tracers were used, where the former included water, iodide, bromide, and lignosulfonate, while the latter was strontium. The data were then fitted to two models. In one model, the solute flowed in a fracture with parallel walls, and its spread was due to dispersion alone. In the second model, the fracture was modeled as a set of parallel, but disconnected, channels and the solute spread was due to only the differences between the flow velocities in different channels, and dispersion was neglected. Assuming a linear sorption isotherm, the first model is described by

$$\begin{aligned} \frac{\partial C_f}{\partial t} + \frac{1}{R_r} V_f \frac{\partial C_f}{\partial x} &= \frac{D_L}{R_r} \frac{\partial^2 C_f}{\partial x^2} - \frac{2}{w} \frac{1}{R_r} D_e \frac{\partial C_p}{\partial z} \Big|_{z=0}, \\ \frac{\partial C_p}{\partial t} &= \frac{1}{K_p \rho_r} D_e \frac{\partial^2 C_p}{\partial z^2}. \end{aligned} \quad (12.45)$$

Here, subscripts *f* and *p* denote the tracers' concentrations in the fracture and the porous matrix, D_L is the longitudinal dispersion coefficient, D_e is the effective diffusivity in the porous matrix, w is the width of the fracture, R_r is a retardation factor given by $R_r = 1 + 2K_p/w$, with K_p being the bulk partition coefficient and ρ_r being the density of the rock. The second model was still described by Eq. (12.45), except that the dispersion term was deleted. The data were accurately represented by both models.

Novakowski *et al.* (1985) and Raven *et al.* (1988) carried out dispersion experiments in a single natural fracture. In the work of the former group, a discrete fracture at a depth of approximately 100 m, located between two boreholes in a relatively horizontal attitude over a distance of about 10 m was used. The experimental technique involved injecting a slug of tracer particles in a steady groundwater flow field established between a pumping and recharging borehole and monitoring the tracer breakthrough by sampling the withdrawal water directly. High injection and withdrawal flow rates were maintained in order to minimize the residence time in both the injection and sampling intervals. Radioactive bromine 82, ^{82}Br , and a fluorescent dye were used as the tracer (solute), and their concentration at the

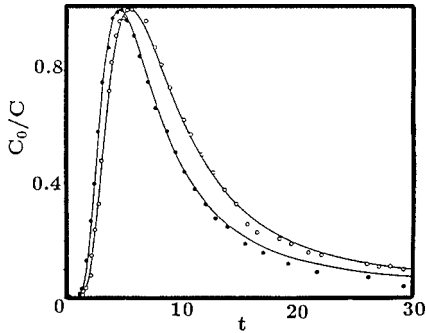


Figure 12.6 Comparison of the experimental data (circles) for normalized concentration C_0/C versus time (in hours) in a single fracture and the predictions of Eq. (11.77) for dispersion in a short porous medium. The two

curves correspond to two different flow rates, and a longitudinal dispersivity of 1.4 m was used in the equation (after Novakowski *et al.*, 1985).

withdrawal borehole was measured. The concentration profile of the tracer was then fitted to the solution of a 1D convective-diffusion (CD) equation for a finite system described in Chapter 11. Figure 12.6 compares the predictions of the fitted model with the experimental data.

Similar experimental studies were carried out by Raven *et al.* (1988). The tracer breakthrough curves were determined from samples of the withdrawn groundwater and were interpreted using the first-passage distributions described in Chapter 11. However, to interpret their data, the authors used a dispersion model similar to the Coats–Smith–Baker model described in Chapter 11, namely, in addition to the usual CD equation, they also used an equation to account for the exchange of the solute between the mobile and stagnant fluids, somewhat similar to Eq. (12.45). They found that such a model is a more accurate representation of their data than a CD equation alone.

Haldeman *et al.* (1991) carried out tracer experiments in a fractured porous tuff block. They then used a 2D model and included the effect of diffusion into the porous matrix, as described by the second equation of Eq. (12.45). However, due to significant channeling, the simulation results did not fit the data. Dronfield and Silliman (1993) attempted to generate a model fracture with rough walls by either inserting obstacles in the space between two parallel plates, or by coating the plates' surface with sand. The data for the tracer experiments were then fitted to a 1D CD equation, from which the longitudinal dispersion coefficient D_L was estimated as a function of the Péclet number Pe (see Chapter 11). They reported that $D_L/D_m \sim Pe^{1.4}$, where D_m is the molecular diffusivity. Note that the exponent 1.4 is larger than that for unfractured porous media which is about 1.2 (see Chapter 11), but smaller than two, the expected exponent for the Taylor–Aris dispersion described in Chapter 11.

Ippolito *et al.* (1994) utilized the echo method to study dispersion in a fracture that consisted of either two flat walls, or one flat wall and a rough one. The rough-

ness was created by acid etching of a zinc plate. In the echo technique, the solution containing the tracer is first injected into the system, and then pumped back through a detector, which reduces the effect of the difference between the flow paths. For Taylor–Aris dispersion in flow between two perfectly flat parallel plates, one has

$$\frac{D_L}{D_m} = 1 + \frac{1}{210} \text{Pe}^2, \quad (12.46)$$

where $\text{Pe} = bV/D_m$, with V being the average flow velocity, and b being the distance between the two plates. In their experiments in a fracture with two flat walls, Ippolito *et al.* (1994) found the same result as Eq. (12.46), except that the numerical factor was $1/217$, perhaps because the walls were not exactly flat and parallel. When one of the walls was rough, Ippolito *et al.* found that the first term on the right side of Eq. (12.46) should be replaced by one proportional to Pe . This may be interpreted as being due to the roughness of the wall, which moves the fracture closer to a porous medium. Recall from Chapter 11 that for dispersion in porous media, D_L is roughly proportional to Pe . The relative importance of the Taylor–Aris dispersion and macrodispersion caused by the flow velocity field was also studied experimentally by Detwiler *et al.* (2000). Boschan *et al.* (2008) studied the same issue using both Newtonian and non-Newtonian fluids.

Keller *et al.* (1999), who studied dispersion of dissolved contaminants in a single fracture, also reported the strong effect of channeling that accelerates the movement of solutes in the direction of channeling, hence resulting in early breakthrough curves. They characterized the fracture and its aperture distribution using computed tomography, and used the dispersion model of Gelhar *et al.* (1979) and Gelhar and Axness (1983) described in Section 11.14.3 to fit their data. One important conclusion was that the geostatistics of a fracture may be used to predict the approximate shape of the breakthrough curve, a conclusion that was also reached by Lee *et al.* (2003) in their experiments with five transparent fractures.

To summarize, extensive experiments indicate that flow channeling in a fracture strongly influences the dispersion process. Clearly, a fracture with a broad distribution of the local apertures also gives rise to a stronger effect of channeling. However, channeling also produces concentration gradients *perpendicular* to the flow direction, which gives rise to molecular diffusion in the transverse direction that, in turn, reduces the effect of the heterogeneities. In addition, diffusion into and sorption in the pores of the porous matrix in which a fracture is embedded is also important.

12.4.2

Asymptotic Analysis

In addition to the theoretical works described in Chapter 11, there have been a few other studies of dispersion in a single fracture with rough internal surfaces. Similar to fluid flow and conduction, we consider a fracture with a rough surface consisting of N segments, where each segment or channel is oriented at some angle θ_i with

respect to the mean plane with an effective aperture, $b_i = \langle b \rangle \cos \theta_i$, and length $\xi_{||}^i = \xi_{||} / \cos \theta_i$ (see Figure 12.1). The roughness profile is assumed to be described by a fractional Brownian motion with the Hurst exponent H . It is also assumed that the segment of length $\xi_{||}$ in the direction of mean flow, over which the channel is formed by the two fracture surface, is approximately straight (see Figure 12.1). The notations in what follows is the same as those for the effective permeability and conductivity, described earlier.

The total length L_t of a channel is given by

$$L_t = \sum_{i=1}^N \xi_{||}^i = \xi_{||} \sum_{i=1}^N \cos^{-1}(\theta_i). \quad (12.47)$$

Thus, the tortuosity factor τ , which is important to diffusion and dispersion in any porous medium, is given by

$$\tau = \frac{L_t}{L} = \langle \cos^{-1}(\theta_i) \rangle \approx 1 + \frac{1}{2} \left(\frac{\sigma_z(\xi_{||})}{\xi_{||}} \right)^2, \quad (12.48)$$

where L is the fracture's length. Here, $\sigma_z^2(\xi_{||})$ is the variance of the segments. If the roughness profile is self-affine, then

$$\sigma_z^2(\xi_{||}) \sim \zeta(l) \left(\frac{\xi_{||}}{l} \right)^{2H}, \quad (12.49)$$

with l being a microscopic length scale. If one assumes that the local heights or apertures in a fracture follow a Gaussian distribution, one obtains a more accurate expression for τ (Drazer and Koplik, 2001),

$$\tau \sim 1 + \left[\frac{\zeta(l)}{l^2} \right] \left(\frac{\langle b \rangle}{l} \right)^{\frac{2H-2}{H}}. \quad (12.50)$$

The effective diffusivity D_e in the fracture is given by $D_e = D_m / \tau^2$, where D_m is the usual bulk molecular diffusivity. Thus, Eq. (12.50) may be recast in terms of the diffusivities,

$$\frac{D_m - D_e}{D_m} \sim 2c \left(\frac{\langle b \rangle}{l} \right)^{\frac{2H-2}{H}}, \quad (12.51)$$

where c is an adjustable parameter of $\mathcal{O}(1)$. Simulations of Drazer and Koplik with $H = 0.8$ yielded results that were in close agreement with Eq. (12.51).

To derive a similar equation for the longitudinal dispersion coefficient D_L , Drazer and Koplik (2001) modified Eq. (12.46) and rewrote it as

$$\frac{D_L}{D_m} = \left(\frac{1}{\tau^2} \right) \left[1 + \frac{\text{Pe}^2}{210} \left(\frac{\langle b \rangle}{\mathcal{A}\tau} \right)^2 \right]. \quad (12.52)$$

Here, $\mathcal{A}$ is the parameter introduced by Eqs. (9.104) and (9.105) in order to relate the effective permeability to the conductivity of a porous medium (recall that the

effective conductivity and diffusivity of a porous medium are directly linked). According to Eq. (12.52), the basic quadratic dependence of the dispersion coefficient D_L on Pe is not modified by the roughness of the internal surface of a fracture, but the numerical coefficient does change from $1/210$ to one that depends on the roughness exponent H . The numerical simulations of Drazer and Koplik (2001) supported Eq. (12.52).

These results are supposedly valid when the flow is very slow – very small Reynolds numbers Re – but nonvanishing. Roux *et al.* (1998) used scaling analysis and a perturbation expansion to study dispersion in a fracture in which the Re for flow was relatively large. Thus, convection was important. Their most important result was that the *local* dispersion coefficient D_l is much smaller than the *global* one, D_L . Moreover, they found that

$$\frac{D_l}{D_L} \sim Pe^{-H}, \quad (12.53)$$

and more interestingly,

$$\frac{D_l}{D_m} \sim Pe^{1-H}. \quad (12.54)$$

Distinguishing the local dispersion coefficient from the global one is important to tracer tests at field scale because in such tests, the measurements can only be carried out *locally* and over a limited range.

12.4.3

Direct Numerical Simulation

Extensive numerical simulations of dispersion in flow through a single fracture with rough internal surfaces have also been carried out over the past two decades using a variety of techniques. Tsang and Tsang (1989), Moreno *et al.* (1988, 1990), and Moreno and Tsang (1991) used a particle-tracking method (see Chapter 11) to study dispersion in a single fracture that was represented by a 2D heterogeneous network, as described earlier for simulating fluid flow. Muradlidhar (1990) simulated dispersion in flow in channels with flat walls in which obstacles had been inserted in a regular pattern. Johns and Roberts (1991) studied fractures with a binary aperture distribution such that a straight rectangular channel was connected to two narrower channels. Convection, diffusion in the flow, diffusion into the matrix, and sorption were all considered. Koplik *et al.* (1993) studied dispersion in a channel between a flat wall and one with a rough surface, and determined the dispersion coefficient. They showed that the roughness greatly increases the dispersion coefficient. Many of such simulations also indicated that the effective dispersion coefficient is sensitive to the precise injection conditions of the tracers.

Plouraboué *et al.* (1998) developed a numerical scheme and lubrication approximation for fluid flow in a fracture. Assuming that the fluid is incompressible, the mass continuity equation implies that $\nabla \cdot \mathbf{v} = 0$. Substituting Darcy's law for the

velocity v then yields

$$\nabla^2 P = -\frac{3}{2} \frac{\nabla K(\mathbf{x})}{K(\mathbf{x})} \cdot \nabla P = -\frac{3}{2} \nabla[\ln K(\mathbf{x})] \cdot \nabla P, \quad (12.55)$$

where the factor $3/2$ is due to the velocity profile for laminar flow between two flat plates. Introducing the stream function $\psi(\mathbf{x})$, one obtains

$$\nabla^2 \psi(\mathbf{x}) = \frac{3}{2} \nabla[\ln K(\mathbf{x})] \cdot \nabla \psi(\mathbf{x}), \quad (12.56)$$

which should be solved with

$$\nabla P(\mathbf{x}) \cdot \nabla \psi(\mathbf{x}) = 0, \quad (12.57)$$

for the two unknowns $P(\mathbf{x})$ and $\psi(\mathbf{x})$. Equation (12.55) also suggests an expansion scheme for the pressure, $\nabla P^{(0)}(\mathbf{x}) + \nabla P^{(1)}(\mathbf{x}) + \dots + \nabla P^{(n)}(\mathbf{x})$. Therefore,

$$\nabla^2 P^{(n)}(\mathbf{x}) = -\frac{3}{2} \nabla[\ln K(\mathbf{x})] \cdot \nabla P^{(n-1)}(\mathbf{x}). \quad (12.58)$$

Thus, the right side of Eq. (12.58) can be computed directly, whereas the left side is calculated in the Fourier space, which allows for a very efficient numerical scheme. Both Plouraboué *et al.* (1998) and Roux *et al.* (1998) identified an anomalous dispersion regime in which the dispersion coefficient D_L depends on the distance traveled from the point of injection of the tracers into the flow.

12.4.4

Lattice-Boltzmann Simulation

Similar to fluid flow in a single fracture, the lattice Boltzmann (LB) technique has also been utilized to study dispersion in fractures with rough internal surfaces. Flekkøy (1993) suggested a LB simulator for flow of miscible fluids and, hence, dispersion that represents the limit in which the two fluids have the same viscosity and density. Drazer and Koplik (2001, 2002) and Madadi and Sahimi (2011) utilized the LB technique to simulate dispersion in flow through a fracture with rough internal surfaces, with the latter group also studying dispersion in a *network* of interconnected fractures with rough internal surfaces (see below). Drazer and Koplik's simulations indicated that dispersion is decreased by convective transport if the fracture surfaces are laterally shifted.

12.5

Flow and Conduction in Fracture Networks

Once fluid flow in a single fracture is understood, fluid flow in a network of interconnected fractures is conceptually straightforward, and analogous to flow in a single pore and its relation with fluid flow in pore networks. Since the connectivity

of the fractures strongly influences the macroscopic properties of the network, the concepts of percolation theory are certainly relevant and, in fact, provide a powerful tool for understanding flow and transport in networks of interconnected fractures. Models of fracture networks were already described in Chapter 8 and, thus, only a brief description of such models is presented, if need be.

Berkowitz and Braester (1991) and Margolin *et al.* (1998) used square and simple-cubic networks in which the bonds represented the fractures. An effective radius or aperture selected from a log-normal distribution was attributed to each bond. Interesting 2D models of fracture networks were developed by Odling and Webman (1991) and Barthélémy *et al.* (1996). They discretized real fracture traces on a square network.

Similar to models of fracture networks described in Chapter 8, Rasmussen *et al.* (1985) used disks in order to generate a 3D model of a fracture network. Each intersection between two fractures represented a node of the network. Two nodes were related if they belong to the same fracture. As such, the model was similar to that of Long *et al.* (1985), already described in Chapter 8.

If the distribution of the fractures is broad, it may be viewed as the superposition of n monoscale elementary lattices, for example, simple-cubic lattices, which represent the different types and scales of fractures in rock. Such a model was developed

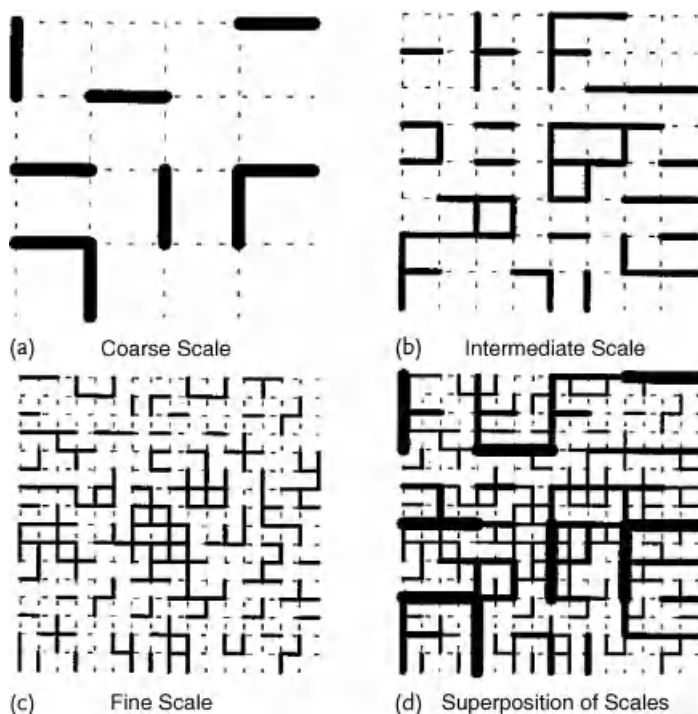


Figure 12.7 A 2D example of a multiscale network of a fracture network. (a)–(c) show three distinct scales, while (d) represents a superposition of the three scales (after Xu *et al.*, 1997).

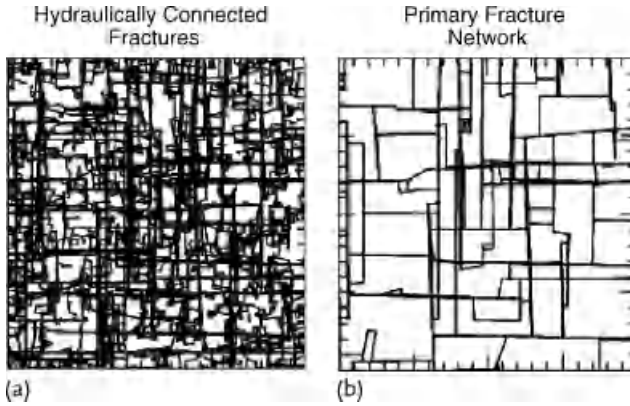


Figure 12.8 Structure of a hierarchical fracture network (after Clemo and Smith, 1997). (a) All the hydraulically-connected fractures. (b) The main fractures that contribute the most to fluid flow.

by Xu *et al.* (1997) (they developed their model for porous media, but it is equally applicable to fracture networks) and Gavrilenko and Guéguen (1998), although Neimark (1989) had already developed such a model in the physics literature. The length L_i and aperture b_i of fracture in lattice i are selected according to a geometric progression of ratio r , $L_i = L_1/r^{i-1}$ and $b_i = b_1/r^{i-1}$, where L_1 and b_1 refer to the largest scale. In each lattice at each scale, the bonds representing the fractures exist with a probability p_i . Such models are called *multiscale networks*. Figure 12.7 shows a 2D example of such networks, indicating the potential utility of such a model in that it contains fractures at several scales.

Clemon and Smith (1997) described another hierarchical model of fracture networks based on the so-called Levy–Lee (LL) model. The LL model generates a fracture network with a continuum of scales for both fracture lengths and the distance between them. The location of the fractures' centers is selected by a random walk. Each new fracture center is at a random distance away from the last fracture. The distance L_c between fractures' centers is selected from a power-law distribution,

$$P_L(L_c > L) \sim L^{-D_f}, \quad (12.59)$$

where D_f is a fractal dimension. The direction of the displacements is uniformly distributed between 0 and 360°. The fracture trace length and the variation in fracture orientation are proportional to the distance from the last inserted fracture. The fracture length follows a power-law distribution. Depending on the value of the fractal dimension D_f , the fractures may be highly clustered. Figure 12.8 presents an example of the type of fracture network generated by the model. The primary fractures are those that dominate the flow behavior of the network.

12.5.1

Numerical Simulations

Once the network is set up, one can compute its flow and conduction properties by methods similar to those used for the pore networks. Such methods include both analytical analysis and numerical techniques. For example, Gavrilenko and Guéguen (1998) used the position-space renormalization group that was described in Chapter 10 in order to compute the flow properties of their multiscale network model.

However, the most comprehensive studies of flow and conduction in networks of interconnected fractures were carried out by Adler, Thovert, and their group (Mourzenko *et al.*, 1997, 1999, 2001, 2004; Koudina *et al.*, 1998; Sisavath *et al.*, 2004; Hamzehpour *et al.*, 2009). Their model, already described in Chapter 8, consists of interconnected identical or polydispersed polygons of various shapes, such as hexagons. They are then discretized and triangulated in order to be used in the simulation of flow and conduction in the fracture network. The triangulation method that they used was also described in Chapter 8. More details of their model and method can also be found in Adler and Thovert (1999). Various power laws for the effective permeability and conductivity of the network near the connectivity or percolation threshold, which were described in Chapter 3, were also studied by this group. For example, the critical exponent μ_p of the conductivity of 3D networks near the percolation threshold (see Chapter 3) was determined by Adler, Thovert and their group. They reported that $\mu_p/\nu \simeq 2.22 \pm 0.08$, which is in agreement with the prediction of percolation theory.

Based on a percolation model of fracture networks, Margolin *et al.* (1998) developed an interesting correlation for the effective permeability K_e in d dimensions if the permeability value K_n measured at any smaller scale is known. Their relation is given by

$$\frac{K_e}{K_n} = \exp(-m), \quad (12.60)$$

where $m = \sigma_{\ln K}^a \ln n / (1 + d)^a$. Here, a and n are two parameters that are determined graphically (see Margolin *et al.*, 1998), and $\sigma_{\ln K}$ is the standard deviation of the log hydraulic conductivity or permeability. We should mention the work of Madadi and Sahimi (2003) who used a lattice-Boltzmann technique in order to simulate fluid flow in networks of interconnected fractures with rough internal surfaces. To our knowledge, their work is the only one in which the surface roughness of all the interconnected fractures was explicitly accounted for in the simulations.

12.5.2

Effective-Medium Approximation

As described in Chapters 6 and 8, fracture networks are often anisotropic and, therefore, they are characterized by a permeability tensor, rather than a single effective permeability. One way of computing the effective permeability tensor is

through the anisotropic EMA (AEMA), described in Section 10.2.8. Its use for fracture networks was first described by Harris (1990, 1992).

We characterize a d -dimensional anisotropic network by d effective permeabilities K_{e_i} with $i = 1, \dots, d$, one for each principal direction. The anisotropy of the network may be caused by several factors. For example, each principal direction may be characterized by its own permeability distribution, as many natural porous media are stratified and, thus, may have distinct distributions of heterogeneities in different directions. Alternatively, the anisotropy may be due to the orientation distribution of the fractures. For example, consider a 2D fracture system that consists of two fracture sets, with the fractures in each set having the same orientation. An example is shown in Figure 12.9. It is clear that this system is anisotropic and is characterized by two effective permeabilities, one for each of its principal directions.

We consider both types of anisotropies and present the results. Consider, first, an anisotropic network in which each principal direction is characterized by its own conductance distribution $f_i(g)$ with $i = 1, \dots, d$. For a simple-cubic network in d -dimensions, the effective conductances g_{e_i} are predicted by the AEMA to be the solutions of Eq. (10.33),

$$\int_0^\infty f_i(g) \frac{g - g_{e_i}}{g + S_i} dg = 0, \quad i = 1, \dots, d. \quad (12.61)$$

The quantity S_1 for a square network is given by Eq. (10.37), while S_2 is obtained by interchanging g_{e_1} and g_{e_2} . The quantity S_1 for a simple-cubic network is given by Eq. (10.38), while S_2 and S_3 are obtained by cyclic rotation of g_{e_1} , g_{e_2} , and g_{e_3} . Observe that in the limit in which all the effective conductance distributions are equal, these equations reduce to an equation for isotropic media, as they should. Once the

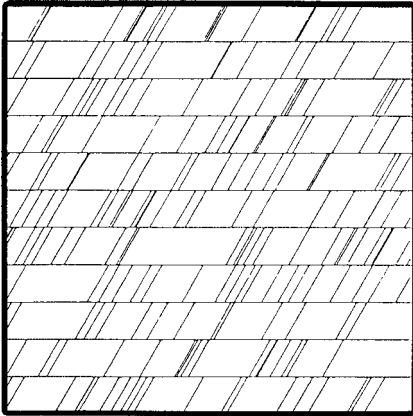


Figure 12.9 A 2D fracture network with two different sets of fractures, each having its own orientation (after Harris, 1990).

effective conductances are calculated, the effective permeabilities are determined by the same method described in Chapter 8 for isotropic media.

Now, consider the second type of anisotropy (Harris, 1990). For concreteness, we consider the fracture network shown in Figure 12.9 that consists of two sets of fractures. One set consists of long, parallel, and equally spaced fractures, whereas the second set is made of those with centers that are randomly and uniformly distributed on the lines parallel to the fracture profiles of the first set. Such a fracture system is mapped exactly onto the network shown in Figure 12.10. Suppose that in the network shown in Figure 12.10, p_i is the probability that a fracture of type i has nonzero (hydraulic) conductance. Suppose also that ζ is the number of the centers of the fracture in the set two per unit length, lying on a line parallel to and midway between a pair of adjacent fractures in set one. Then,

$$p_2 = \zeta \ell_2, \quad p_1 = 1, \quad (12.62)$$

where ℓ_2 is the length of the fractures in set two (see Figure 12.10). The network shown in Figure 12.10 is anisotropic, not only because of the orientations of the fractures, but also due to the fact that the fractures in set one and set two have different conductance distributions, as their lengths are very different. However, since the network is still four-coordinated, and the angles that the fractures make with each other are the same everywhere, Eqs. (10.37) and (10.38) can be used. The two conductance distributions are then given by $f_1(g) = h_1(g)$ and $f_2(g) = (1 - p_2)\delta(g) + p_2 h_2(g)$, where $h_1(g)$ and $h_2(g)$ are two normalized probability distributions that describe the conductances of the fractures in set one and set two. Note that because $p_1 = 1$, $f_1(g)$ does not contain any $\delta(g)$. Using Eqs. (10.37) and (10.38), one can show that

$$\tan^2 \left[\frac{1}{2} \pi (1 - p_2) \right] \leq \frac{g_{e1}}{g_{e2}} \leq \tan^2 \left(\frac{1}{2} \pi p_1 \right). \quad (12.63)$$

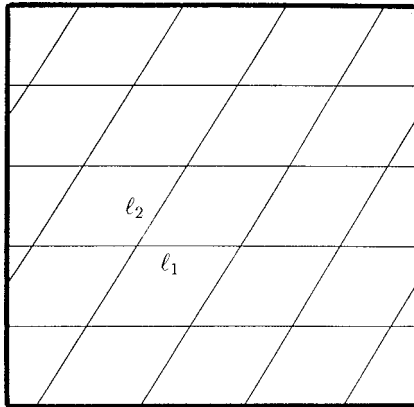


Figure 12.10 The equivalent network of the fracture network shown in Figure 12.9 (after Harris, 1990).

Thus, the idea is to map the fracture network onto an equivalent regular network, for which the necessary formulations for constructing the EMA and AEMA (see Chapter 10) are already available. Harris (1990, 1992) worked out several examples, and Mukhopadhyay and Sahimi (2000) derived the general AEMA for an arbitrary regular network which was described in Chapter 10.

12.6

Dispersion in Fracture Networks

To study dispersion in a network of fractures, the problem must first be understood at two different levels of complexities. The first one is dispersion in flow through a single fracture. This was already studied in detail in Section 12.4. At the next level of complexity is dispersion at the intersection of two or more fractures. It has been assumed in most of the previous works that the solute and solvent mix completely at the fracture intersections, similar to a class of model developed by Sahimi and co-workers (Sahimi *et al.*, 1983a, 1986a,b; Sahimi and Imdakm, 1988) for dispersion in pore networks that we described in Chapter 11. The assumption of complete mixing at the fracture intersections is partly based on the works of Castillo *et al.* (1972) and Krizek *et al.* (1972), who used a plexiglass model of two intersecting fractures. Their work indicated that the assumption of complete mixing at the intersections was justified. However, Hull and Koslow (1986) pointed out that the older works were not conclusive.

On the other hand, Wilson and Witherspoon (1975, 1976) reported that there is little or no mixing at the fracture intersections. Thus, Endo *et al.* (1984) studied dispersion in a fracture network in which *no* mixing was assumed at the intersections. The laboratory work of Hull and Koslow (1986) and Robinson and Gale (1990), using discrete networks of fractures, indicated that the assumption of complete mixing at the fracture intersections is not justified. Although there is some mixing at the intersections, its intensity depends on the morphology of the fracture network and the flow regime in it. For example, if the flow field is slow enough, diffusive mixing may occur. Note that what happens at the intersection of the fractures has a strong influence on the distribution of the solute in the fracture network. Figure 12.11 shows two scenarios for dispersion in a fracture intersection. In one, complete mixing of the solute and solvent is assumed, whereas in the second, no mixing is assumed. It should be clear that the two networks have very different distribution of the solute particles. Detailed simulations of the problem of solute partitioning and mixing at the fractures' intersections were carried out by Mourzenko *et al.* (2002). Once dispersion at the level of a single fracture and at the intersection of two or more fractures is understood, one can study dispersion in a network of fractures.

In most of fracture networks models that have been used so far, dispersion is simulated by a random walk or particle-tracking method described in Chapter 11 for pore networks and for field-scale porous media. The only exception that we are

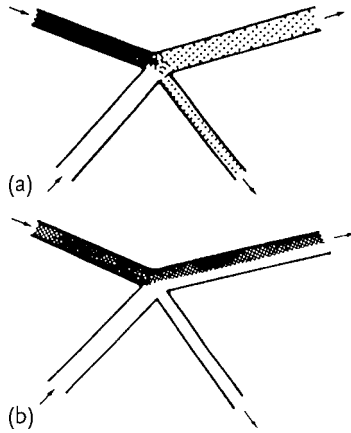


Figure 12.11 Two scenarios for dispersion at an intersection of fractures: complete mixing (a) and no mixing (b) (after Cacas *et al.*, 1990b).

aware of is the work of Madadi and Sahimi (2011) who used the lattice-Boltzmann technique in order to simulate dispersion in flow through a network of interconnected fracture, each having a self-affine rough internal surface.

As an example of the typical simulation of dispersion in flow through a network of interconnected fracture, consider the early work of Schwartz *et al.* (1983) and Smith and Schwartz (1984). They used 2D networks of interconnected discrete fractures – models that were already described in Chapter 8. The fractures were oriented at various angles with respect to the direction of the macroscopic mean flow. After calculating the flow field throughout the network, tracer (solute) particles were released into the network and their motion, which was governed by a random walk biased by the macroscopic mean flow (see Chapter 11), was monitored. The breakthrough curves were computed, and the question of whether the curves represented a Gaussian distribution was studied.

Using such network models and simulation techniques, dispersion in flow through fracture networks has been studied extensively. Such studies, in addition to the work of Schwartz *et al.* (1983) and Smith and Schwartz (1984), include those of Endo *et al.* (1984), Rasmuson (1985), Hull *et al.* (1987), Robinson and Gale (1990), Cacas *et al.* (1990a,b), Tsang *et al.* (1991), Berkowitz and Braester (1991), Moreno and Neretnieks (1993a,b), Clemo and Smith (1997), Margolin *et al.* (1998), and Huseby *et al.* (2001), utilizing various types of discrete networks of interconnected fractures.

In particular, Berkowitz and Braester (1991) utilized square networks in which the apertures were distributed according to a log-normal distribution. Using the random walk method (see Chapter 10), they studied dispersion in the fracture network. The mean-square displacement of the solute tracers was found to follow

$$\langle R^2(t) \rangle \sim t^{1.27}, \quad (12.64)$$

in excellent agreement with the prediction of Sahimi (1987), Eq. (11.89), that yields $\langle R^2(t) \rangle \sim t^{1.26}$. On the other hand, Moreno and Neretnieks (1993a,b) used simple-cubic networks together with a random walk method similar to that of Sahimi *et al.* (1983a, 1986a,b, see Chapter 11) to study dispersion of a contaminants in flow through a fracture network. Margolin *et al.* (1998) utilized their anisotropic percolation model of fracture networks mentioned earlier to study solute transport and obtain a relation between the variations in the apertures and the Péclet number.

Detailed simulations of dispersion in flow through fracture networks were carried out by Huseby *et al.* (2001) using the model developed by Adler, Thovert and their group described in Chapter 8. Interestingly, they reported that both the longitudinal and transverse dispersion coefficients depended on the Péclet number Pe as a power law with the *same* exponent, $D_L \sim Pe^\alpha$ and $D_T \sim Pe^\alpha$, with the interesting twist that α appeared to depend on the density of the fractures, ranging from about 1.75 at low densities to about 1.1 for the high values. The high value of α at low fracture densities may be understood by recognizing that dispersion in a low-density fracture network in which the fractures rarely intersect one another is close to the Taylor–Aris dispersion, for which $D_L \sim Pe^2$. On the other hand, in a high-density network, it is the velocity field that dominates dispersion, hence the low values of α close to those for disordered porous media described in Chapter 11.

On the experimental side, Hull *et al.* (1987) and Robinson and Gale (1990) constructed 2D networks of fractures, and carried out dispersion experiments in them. They also performed random walk simulations of dispersion in similar networks in order to test the validity of the assumptions that they made in their models.

In most cases, it has been found that the most important controlling factors are the orientation of the fracture sets with respect to the macroscopic mean flow, and the density of the fractures in a given region of the network. Moreover, the distribution of the apertures has a strong influence on the distribution of the solute particles throughout the fracture network. Thus, uncertainties in predicting the solute concentration profile is larger in networks with larger variability in their aperture distribution.

One of the most important results of studies of dispersion in a network of fractures is that in most cases, dispersion may *not* be Gaussian or diffusive. As discussed in Chapter 6, several careful experimental studies have indicated that fracture networks of rock may have a fractal, scale-invariant structure. If so, dispersion in such networks should be characterized by dispersivities and dispersion coefficients that vary with time and length scales, similar to those described in Chapter 11 for field-scale porous media. Even though most of the networks used in the Monte Carlo simulations of dispersion in flow through fracture networks were not fractal, the fact that dispersion is found to be nondiffusive indicates its complexity in fracture networks and fractured porous media. On the other hand, assuming that a fracture network is a fractal object, Ross (1986) provided some arguments about the variations of the dispersivities with the scale of observations and the distance traveled.

12.7

Flow and Transport in Fractured Porous Media

Flow in fractured porous media has been studied by both a continuum approach and by network models. The continuum approach has relied heavily on the so-called double-porosity model, which the petroleum industry has made extensive use of. Network models can be studied by an EMA, a generalization of what we described in Chapter 10 for unfractured porous media, and by computer simulations. As usual, we describe both approaches in an attempt to understand their strengths and shortcomings.

12.7.1

The Double- and Triple-Porosity Models

To understand the basis for the double-porosity models, consider first a single-porosity system – an unfractured porous medium. Suppose that the fluid is compressible with a compressibility factor c . Then, an equation of state for the fluid is usually written as (see, for example, Muskat, 1937)

$$\rho_f = \rho_0 \left(\frac{P}{P_0} \right)^m \exp[c(P - P_0)] , \quad (12.65)$$

where ρ_f is the density of the fluid, P is the pressure, and ρ_0 and P_0 is some reference state. The value of the parameter m depends on the fluid. For liquids, $m = 0$, while for gases, $m = 1$ for isothermal processes, and $m = C_V/C_p$ for adiabatic processes, where C_V and C_p denote, respectively, the specific heat at constant volume and constant pressure. We restrict our attention to liquids for which $m = 0$. If we assume that the liquid is only slightly compressible, we may expand the exponential term in Eq. (12.65) and only keep the first term to obtain

$$\rho_f \simeq \rho_0 + c\rho_0(P - P_0) . \quad (12.66)$$

On the other hand, the equation of continuity, using Darcy's law, is given by

$$\phi \frac{\partial \rho_f}{\partial t} = \nabla \cdot \left(\frac{\rho_f K}{\mu} \nabla P \right) , \quad (12.67)$$

where ϕ is the porosity, μ is the viscosity, and K is the permeability that varies spatially. Substituting Eq. (12.66) into Eq. (12.67) yields

$$\frac{K}{\mu} \nabla^2 P = c\phi \frac{\partial P}{\partial t} . \quad (12.68)$$

The basis for the double-porosity model is to generalize Eq. (12.68) to a system of fractures and pores. Thus, if subscripts one and two denote the pore and fracture systems, respectively, Barenblatt and Zheltov (1960) proposed the following set of

equations for describing flow of a slightly compressible fluid through a system of fractures and pores,

$$\frac{K_1}{\mu} \nabla^2 P_1 - \frac{\alpha}{\mu} (P_1 - P_2) = c_1 \phi_1 \frac{\partial P_1}{\partial t}, \quad (12.69)$$

$$\frac{K_2}{\mu} \nabla^2 P_2 + \frac{\alpha}{\mu} (P_1 - P_2) = c_2 \phi_2 \frac{\partial P_2}{\partial t}, \quad (12.70)$$

where α is a characteristic of the porous medium that has the units $1/(\text{length})^2$. The second terms on the left sides of Eqs. (12.69) and (12.70) represent the *exchange* between the fractures and the porous matrix if it is assumed that the fluid exchange is a pseudo-steady-state process. This assumption can be relaxed, and is discussed further below. Note that such a formulation assumes that the fractures and porous matrix can be characterized by the permeabilities K_1 and K_2 and porosities ϕ_1 and ϕ_2 . If there are large-scale variations of the permeabilities, as in field-scale porous media, then Eqs. (12.69) and (12.70) should be written in more general forms to allow for the spatial variations of the permeabilities. Thus, they are rewritten as (see Eq. (12.67))

$$\nabla \cdot (K_1 \nabla P_1) - \alpha (P_1 - P_2) = c_1 \mu \phi_1 \frac{\partial P_1}{\partial t}, \quad (12.71)$$

$$\nabla \cdot (K_2 \nabla P_2) + \alpha (P_1 - P_2) = c_2 \mu \phi_2 \frac{\partial P_2}{\partial t}. \quad (12.72)$$

If we assume that, compared with K_2 , K_1 is negligible and that the porosity ϕ_2 of the fractures is also negligible compared with ϕ_1 , the porosity of the matrix, then it is not very difficult to show that

$$\frac{\partial P_i}{\partial t} - \frac{K_2}{\alpha} \frac{\partial}{\partial t} [\nabla \cdot (\nabla P_i)] - \frac{K_2}{c_1 \phi_1 \mu} \nabla \cdot (\nabla P_i) = 0, \quad i = 1, 2. \quad (12.73)$$

In the limit $\alpha \rightarrow \infty$, Eq. (12.73) reduces to the classical diffusion equation that describes the flow of a slightly compressible fluid in a porous medium with K_2 and ϕ_1 as its permeability and porosity. Equation (12.73) was first given by Barenblatt *et al.* (1960). The celebrated Warren–Root model (Warren and Root, 1963) of naturally fractured reservoirs represents this limiting case, except that Warren and Root did not neglect the storage capacity of the fractures. Thus, their model is described by

$$\alpha (P_2 - P_1) = c_1 \mu \phi_1 \frac{\partial P_1}{\partial t}, \quad (12.74)$$

$$K_2 \nabla \cdot (\nabla P_2) + \alpha (P_1 - P_2) = c_2 \mu \phi_2 \frac{\partial P_2}{\partial t}. \quad (12.75)$$

The solution of the double-porosity model is usually obtained for a configuration of the system in which one or more wells have been drilled in the reservoir. Then, the main goal of the model is to calculate the pressure at the well(s). This is usually called *well-testing*, and is a standard method of gaining information on the structure of the reservoir since the solutions of Eqs. (12.74) and (12.75), which contain

characteristic parameters such as the permeability and porosity, can be fitted to experimental data, from which the permeabilities and porosities may be estimated. For example, the solution of the problem for an axisymmetric system (that is, in cylindrical coordinates without the angular or axial dependence (see Chapter 2)) can be obtained. We introduce dimensionless quantities defined by

$$P_{di} = \frac{2\pi K_2 h (P_0 - P_i)}{\mu Q}, \quad i = 1, 2 \quad (12.76)$$

where h is thickness of the reservoir, and Q is the flow rate of the well at which the response of the reservoir is measured,

$$r_d = \frac{r}{r_w}, \quad (12.77)$$

where r_w is the radius of the well and

$$t_d = \frac{K_2 t}{\mu r_w^2 (c_1 \phi_1 + c_2 \phi_2)}, \quad (12.78)$$

$$\Phi = \frac{c_2 \phi_2}{c_1 \phi_1 + c_2 \phi_2}, \quad (12.79)$$

$$z = \frac{\alpha r_w^2}{K_2}. \quad (12.80)$$

In dimensionless forms, Eqs. (12.74) and (12.75) are then given by

$$z(P_{d2} - P_{d1}) = (1 - \Phi) \frac{\partial P_{d1}}{\partial t_d}, \quad (12.81)$$

$$\frac{1}{r_d} \frac{\partial}{\partial r_d} \left(r_d \frac{\partial P_{d2}}{\partial r_d} \right) + z(P_{d1} - P_{d2}) = \Phi \frac{\partial P_{d2}}{\partial t_d}. \quad (12.82)$$

The initial and boundary conditions are $P_{d2}(\infty, t_d) = 0$ if the reservoir is of infinite extent, and

$$\frac{\partial P_{d2}}{\partial r_d} \Big|_{r_{de}} = 0 \quad (12.83)$$

if it is of finite extent, where r_{de} is the external radius of the reservoir (hence, Eq. (12.83) is simply the zero-flux condition on the boundary of the reservoir). Moreover,

$$\frac{\partial P_{d2}}{\partial r_d} \Big|_{r_d=1} = -1 \quad (12.84)$$

and $P_{d1}(r_d, 0) = P_{d2}(r_d, 0) = 0$. Warren and Root (1963) obtained an approximate solution of this boundary-value problem in the Laplace transform space. If λ is the Laplace transform variable conjugate to t , then the Warren–Root solution in the Laplace transform space is given by

$$\tilde{P}_{d2} = \frac{Y_0 \left[r_d \sqrt{\lambda f(\lambda)} \right]}{\lambda \sqrt{\lambda f(\lambda)} Y_1 \left[\sqrt{\lambda f(\lambda)} \right]}, \quad (12.85)$$

with

$$f(\lambda) = \frac{\Phi(1 - \Phi)\lambda + z}{(1 - \Phi)\lambda + z},$$

where Y_i is the i th-order modified Bessel function of the second kind.

Note that according to the solution of the Warren–Root model, two parameters, namely, Φ , which is the ratio of the storage capacity of the fractures and the total storage capacity of the reservoir, and z , which is related to the permeability of the fractures, suffice to characterize the behavior of fluid flow in a naturally fractured reservoir. Figure 12.12 displays typical Warren–Root solutions for an infinite reservoir and some values of Φ and z . Bourdet *et al.* (1983) proposed that in order to distinguish between various types of reservoir, one should look at a plot of $\partial P_d / \partial (\log t_d) = t_d \partial P_d / \partial t_d$ versus t_d . Typical behavior obtained in this way is shown in Figure 12.13. For a homogeneous, single-porosity porous medium, the curve should become horizontal and, thus, a plot like Figure 12.13 is characteristic of a fractured reservoir.

Fluid exchange between the matrix and fractures is not necessarily a pseudo-steady-state process. One may argue that the exchange between the two systems is a transient phenomenon and, thus, the terms that represent it should be proportional to the gradient of the pressure at the interface between the fracture and matrix, rather than the difference between the pressures in the bulk of the two systems, as assumed above. Such an idea was first proposed by Kazemi (1969), who also obtained the solution of the problem for a relatively-simple case.

One may generalize the double-porosity model to a triple-porosity one, or even one in which one has four or more distinct porosities and permeabilities that char-

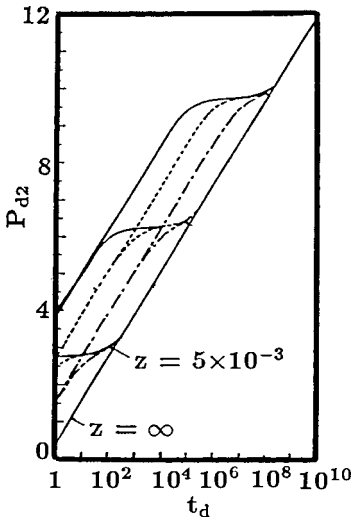


Figure 12.12 Typical solutions of the Warren–Root model for an infinite reservoir. The inclined curves are, from top to bottom, for $\Phi = 0.001, 0.01, 0.1$, and 1 . The two upper horizontal curves are for $z = 5 \times 10^{-6}$ and 5×10^{-9} (after Warren and Root, 1963).

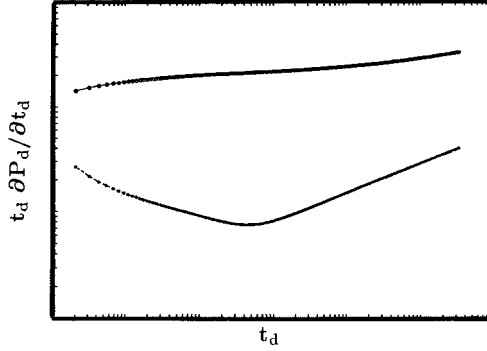


Figure 12.13 Typical time-dependence of the derivative of pressure for a double-porosity porous medium. For comparison, the behavior of the pressure (upper curve) is also shown.

acterize the reservoir or rock. The triple-porosity model was first proposed by Closmann (1975). In his model, one has two types of porous media, one with good flow properties, and a second one with poor properties. The third degree of porosity is contributed by the fractures. It was proposed that such a model may be appropriate for describing fractured carbonate reservoirs. If subscript three denotes the fractures, and one and two denote the two porous matrices, then a triple-porosity model is expressed by the following equations

$$K_1 \nabla^2 P_1 + \alpha_1 (P_3 - P_1) = c_1 \mu \phi_1 \frac{\partial P_1}{\partial t} , \quad (12.86)$$

$$K_2 \nabla^2 P_2 + \alpha_2 (P_3 - P_2) = c_2 \mu \phi_2 \frac{\partial P_2}{\partial t} , \quad (12.87)$$

$$K_3 \nabla^2 P_3 - \alpha_1 (P_3 - P_1) - \alpha_2 (P_3 - P_2) = c_3 \mu \phi_3 \frac{\partial P_3}{\partial t} . \quad (12.88)$$

Closmann (1975) assumed that, relative to K_3 , K_1 and K_2 are negligible. Abdassah and Ershaghi (1986) modified Closmann's model by assuming that the exchange terms are proportional to the pressure gradients between various systems, as had been done by Kazemi (1969) for the double-porosity model. Such models may also be modified further in order to take into account the effect of stratification in the reservoir.

As formulated, the double- and triple-porosity models basically represent the solution of a set of coupled transient diffusion equations, with the coupling terms representing the fluid exchange between the matrix and fractures. Thus, depending upon the initial and boundary conditions, one may obtain a variety of solutions. It is impossible to give a list of all the analytical solutions that have been obtained so far, as the list is too long. The interested reader should consult (Chen, 1989, 1990) for a comprehensive list of references and a more complete discussion.

The double-porosity models (and the triple-porosity ones) have provided a better understanding of flow in a fractured porous medium. They have also been extended to the so-called *double-permeability* models (see, for example, Clemo and Smith,

1989) in which both the fracture network and the porous matrix contribute to the fluid flow and transport. Recall that in the double-porosity model, the matrix is disconnected, and flow and transport occur through the fracture network. They suffer, however, from several shortcomings, the most serious of which is the fact that they do not include any information about the morphology of the matrix and fractures. As we have emphasized throughout this book, the morphology of a porous medium – the connectivity of the pores and fractures, the pore size and aperture distributions, and the fracture density in various regions of the system – are all important parameters that have strong influence on the overall behavior of the fluid flow. Continuum double- or triple-porosity models ignore all of these and represent the matrix and fractures by effective permeabilities and porosities. Given that a fracture network of naturally fractured rock is a fractal object (see Chapter 6), such a representation is inadequate. To take such effects into account, one must resort to network models.

12.7.2

Network Models: Exact Formulation and Perturbation Expansion

Exact formulation of a network model of transport in a multiporosity system was developed by Hughes and Sahimi (1993a,b). The starting point of the analysis is Eqs. (12.71) and (12.72), the set of coupled diffusion equations in which the permeabilities vary spatially. This set is then discretized to obtain the network model of transport in the multiporosity system. The difference between network and continuum models is that in the former model, the topology of the fracture network and the matrix, and the distributions of the matrix permeability and fracture network are explicitly included. Thus, in analogy with the formulation of the problem of transport in pore networks described in Chapter 10, consider the following set of coupled discretized diffusion equations, which is the natural generalization of the familiar master equation on a d -dimensional network described in Chapter 10. To accommodate the existence of N distinct families of transport paths (and porosities), the set must contain N coupled equations:

$$\frac{\partial \mathbf{P}_i(t)}{\partial t} = \sum_j \mathbf{W}_{ij} [\mathbf{P}_j(t) - \mathbf{P}_i(t)] + \mathbf{E}_i \mathbf{P}_i(t). \quad (12.89)$$

Here, $\mathbf{P}_i(t)$ is a column vector, the dimension of which corresponds to the number of distinct transport paths (e.g., pores and fractures). Consistent with the probabilistic interpretation of a discretized diffusion equation given in Chapter 10, the s th component of $\mathbf{P}_i(t)$ represents the probability that at time t , a randomly-moving “particle” is found in path s at network site i . The *transition matrix* $\mathbf{W}_{ij}$ governs the rate at which the bond joining sites i and j is crossed. The ss' entry of the matrix represents the rate at which particles in path s' at site j move to path s at site i , and is related to the permeabilities or hydraulic conductances at sites or grid points i and j . The *exchange matrix* $\mathbf{E}_i$ represents the instantaneous rate of transition be-

tween transport paths at site i , or the rate of exchange between distinct transport paths.

If (1) the underlying network is translationally invariant; (2) the transition matrix $\mathbf{W}_{ij}$ is a function only of the relative positions of sites i and j , and (3) the exchange matrix $\mathbf{E}_i$ is independent of position, Eq. (12.89) is easily solved by Fourier analysis. To illustrate the more general situation, consider a 1D system with two paths for which the transitions between sites are restricted to nearest-neighbor sites. Therefore, for $j = i \pm 1$,

$$\mathbf{W}_{ij} = \begin{pmatrix} \alpha_{ij} & 0 \\ 0 & \beta_{ij} \end{pmatrix} \quad \text{and} \quad \mathbf{E}_i = \begin{pmatrix} -\mu_i & \nu_i \\ \mu_i & -\nu_i \end{pmatrix}. \quad (12.90)$$

In this case, Eq. (12.89) is equivalent to two scalar equations that are spatially discretized versions of continuum equations of double diffusion in one space dimension described earlier.

As the model of a disordered porous medium with N distinct families of transport paths, we make the following assumptions.

1. All network sites are topologically equivalent. The set of nearest neighbors of site i is denoted by $\{i\}$, so that $j \in \{i\}$ means that site j is a nearest neighbor of site i . The coordination number or connectivity of the network is denoted by Z .
2. The transition matrices $\mathbf{W}_{ij}$ are nonzero only when the sites i and j are nearest neighbors.
3. The transition matrices $\mathbf{W}_{ij}$ are independent, identically distributed random variables.
4. The exchange matrices $\mathbf{E}_i$ are independent, identically distributed random variables.
5. The $\mathbf{W}_{ij}$ and $\mathbf{E}_k$ matrices are mutually independent. It is *not* assumed that for a given matrix $\mathbf{W}_{ij}$ or $\mathbf{E}_k$, the individual entries are independent. Assumptions 3–5 may be relaxed, but are imposed here for simplicity.

The explicit solution of the set Eq. (12.89) for a given realization of the disorder is unlikely to be available (except in one dimension) and, indeed, is not of particular interest. We require a statistical characterization of the large-scale effective transport properties of the system. In one dimension, some exact results can be derived (see below), but apart from this case, an exact solution of the problem is unlikely. We develop an exact but implicit solution for matching the disordered medium to a uniform one with an effective transition matrix that is the same for all the bonds and an effective exchange matrix that is the same for all the sites. A discussion of the physical interpretation of the exact solution in the case of a single family of transport paths – an unfractured porous medium – was given in Chapter 10. In what follows, the diagonal matrix with diagonal elements (in order) $\rho_1, \rho_2, \dots, \rho_N$ is denoted by $\text{diag}\{\rho_1, \rho_2, \dots, \rho_N\}$ or, more briefly, by $\text{diag}\{\rho_n\}$.

We attempt to match the disordered multiporosity medium to an “equivalent” uniform one. As described in Chapter 10, the matching procedure is performed in

the Laplace transform space, in which case Eq. (12.89) becomes

$$\lambda \tilde{P}_i - P_i(0) = \sum_{j \in \{i\}} \mathbf{W}_{ij} [\tilde{P}_j(t) - \tilde{P}_i(t)] + \mathbf{E}_i \tilde{P}_i. \quad (12.91)$$

Equation (12.91) is to be matched to

$$\lambda \tilde{P}_i^0 - P_i(0) = \sum_{j \in \{i\}} \tilde{\mathbf{W}}_e(\lambda) [\tilde{P}_j^0(\lambda) - \tilde{P}_i^0(\lambda)] + \tilde{\mathbf{E}}_e \tilde{P}_i^0, \quad (12.92)$$

that represents the uniform system. Similar to the analysis in Chapter 10, the effective transition (conductance) matrix $\tilde{\mathbf{W}}_e$ and the effective exchange matrix $\tilde{\mathbf{E}}_e$ are functions of the Laplace transform variable λ . Hence, in the time domain, the disordered system is actually being matched to a *generalized master equation* with N distinct families of transport paths:

$$\begin{aligned} \frac{\partial P_i^0(t)}{\partial t} &= \int_0^t \sum_{j \in \{i\}} \mathbf{W}_e(t-\tau) [P_j^0(\tau) - P_i^0(\tau)] d\tau \\ &\quad + \int_0^t \mathbf{E}_e(t-\tau) P_i^0(\tau) d\tau, \end{aligned} \quad (12.93)$$

where $\mathbf{W}_e(t)$ and $\mathbf{E}_e(t)$ are *memory kernels*. Based on the analysis presented in Chapter 10, we know that when transport is confined to a fractal subset of the network, the memory kernels can be slowly decaying, leading to fractal (subdiffusive) transport in which the mean-square displacement of a diffusing particle in the network grows more slowly than linearly with time. This is particularly important since as the analysis in Chapter 6 indicated, fracture network of natural rock is often a fractal object. In that case, only a formulation of the problem of the type developed here can take such important features into account. The analog of Eq. (12.93) for a continuum is given by

$$\frac{\partial P}{\partial t} = \int_0^t \mathbf{K}_e(t-\tau) \nabla^2 P(\mathbf{x}, \tau) d\tau + \int_0^t \mathbf{E}_e(t-\tau) P(\mathbf{x}, \tau) d\tau, \quad (12.94)$$

where $\mathbf{x}$ is a point in space. According to Eq. (12.94), matching a heterogeneous continuum to a homogeneous one induces memory. Equation (12.94) should be compared with Eqs. (12.69) and (12.70), or with Eqs. (12.66)–(12.68), which contain no memory.

Since most of the equations are derived in the Laplace transform space, we delete the tilde sign for the memory kernels and use $\mathbf{W}_e(\lambda)$ and $\mathbf{E}_e(\lambda)$. The initial condition is removed by subtracting Eqs. (12.91) and (12.92). After a little rearrangement, we obtain

$$\begin{aligned} &(\mathbf{ZI} + \mathbf{A}) [\tilde{P}_i(\lambda) - \tilde{P}_i^0(\lambda)] - \sum_{j \in \{i\}} [\tilde{P}_j(\lambda) - \tilde{P}_j^0(\lambda)] \\ &= - \sum_{j \in \{i\}} \Delta_{ij} [\tilde{P}_j(\lambda) - \tilde{P}_j^0(\lambda)] + \mathbf{\Gamma}_i \tilde{P}_i(\lambda), \end{aligned} \quad (12.95)$$

where

$$\mathbf{A} = (\mathbf{W}_e)^{-1}(\lambda \mathbf{I} - \mathbf{E}_e) , \quad (12.96)$$

$$\Delta_{ij} = (\mathbf{W}_e)^{-1} \mathbf{W}_{ij} - \mathbf{I} , \quad (12.97)$$

$$\boldsymbol{\Gamma}_i = (\mathbf{W}_e)^{-1}(\mathbf{E}_i - \mathbf{E}_e) . \quad (12.98)$$

The *matrix* Green function $\mathbf{G}_{ij}(\mathbf{A})$ for the difference operator on the left side of Eq. (12.95) satisfies

$$(\mathbf{Z}\mathbf{I} + \mathbf{A})\mathbf{G}_{ik}(\mathbf{A}) - \sum_{j \in \{i\}} \mathbf{G}_{jk}(\mathbf{A}) = -\delta_{ik}\mathbf{I} . \quad (12.99)$$

Equation (12.95) has the formal solution

$$\begin{aligned} \tilde{P}_i(\lambda) - \tilde{P}_i^0(\lambda) &= \sum_j \sum_{k \in \{j\}} \mathbf{G}_{ij}(\mathbf{A}) \Delta_{jk} [\tilde{P}_j(\lambda) - \tilde{P}_k(\lambda)] \\ &\quad - \sum_j \mathbf{G}_{ij}(\mathbf{A}) \boldsymbol{\Gamma}_j \tilde{P}_j(\lambda) , \end{aligned} \quad (12.100)$$

which is exact, but only determines $\tilde{P}_i(\lambda)$ implicitly. It does, however, express the fluctuations in the solution of the disordered medium from that of the uniform one as a sum over the fluctuations in the transition matrices $\mathbf{W}_{ij}$ (via Δ_{jk}) and the exchange matrices $\mathbf{E}_i$ (via $\boldsymbol{\Gamma}_i$). The formal solution is also a convenient starting point for the construction of the EMAs. We will momentarily defer the discussion of the implementation of the EMA to assemble some properties of the uniform system described by Eq. (12.82), which are needed to translate the EMA predictions for $\mathbf{W}_e(\lambda)$ and $\mathbf{E}_e(\lambda)$ into observable quantities. They also reveal some subtleties that arise in the exact analysis in one space dimension.

For d -dimensional regular networks, the site index i is replaced by a vector $\mathbf{r}$ with integer entries. The Green function is constructed by discrete Fourier analysis, and we find in particular that the Green function at the origin, which we need for constructing an EMA, is $\mathbf{G}_{00}(\mathbf{A}) = -\mathbf{G}(\mathbf{A})$, where

$$\mathbf{G}(\mathbf{A}) = \frac{1}{(2\pi)^d} \int_{-\pi}^{\pi} \dots \int_{-\pi}^{\pi} \{Z[1 - \mathcal{A}(\mathbf{k})]\mathbf{I} + \mathbf{A}\}^{-1} d^d \mathbf{k} , \quad (12.101)$$

and

$$\mathcal{A}(\mathbf{k}) = Z^{-1} \sum_{\mathbf{r} \in \{\mathbf{0}\}} \exp(i\mathbf{k} \cdot \mathbf{r}) . \quad (12.102)$$

We introduce a matrix $\mathbf{H}$ to diagonalize $\mathbf{A}$. Thus,

$$\mathbf{H}^{-1}\mathbf{A}\mathbf{H} = \text{diag}\{a_n\} . \quad (12.103)$$

One example of $\mathbf{H}$ is given below. Then,

$$\mathbf{G}(\mathbf{A}) = \mathbf{H} \text{diag}\{g(a_n)\} \mathbf{H}^{-1}, \quad (12.104)$$

where

$$g(a) = \frac{1}{(2\pi)^d} \int_{-\pi}^{\pi} \cdots \int_{-\pi}^{\pi} \frac{d^d \mathbf{k}}{Z[1 - \mathcal{A}(\mathbf{k})] + a} \quad (12.105)$$

is the value at the origin of the (scalar) lattice Green function introduced in Chapter 10. In particular, in one dimension, we have

$$g(a) = \frac{1}{\sqrt{a(4+a)}}. \quad (12.106)$$

The Laplace transform $\langle \tilde{\mathbf{R}}^2(\lambda) \rangle$ of the column vector mean-square displacement

$$\langle \mathbf{R}^2(t) \rangle = \sum_{\mathbf{r}} |\mathbf{r}|^2 P_{\mathbf{r}}(t) \quad (12.107)$$

is $-\nabla_{\mathbf{k}}^2 \hat{P}(\mathbf{k}, \lambda)|_{\mathbf{k}=\mathbf{0}}$, where

$$\hat{P}(\mathbf{k}, \lambda) = \sum_{\mathbf{r}} \exp(i\mathbf{k} \cdot \mathbf{r}) \tilde{P}_{\mathbf{r}}(\lambda), \quad (12.108)$$

is the discrete Fourier transform of $\tilde{P}_{\mathbf{r}}(\lambda)$. For the initial condition,

$$P_{\mathbf{r}}(0) = \delta_{\mathbf{r}\mathbf{0}} \mathbf{u}, \quad (12.109)$$

taking the discrete Fourier transform of Eq. (12.92), we deduce that

$$\hat{P}(\mathbf{k}, \lambda) = \mathbf{F}(\mathbf{k}, \lambda)^{-1} \mathbf{u}, \quad (12.110)$$

where

$$\mathbf{F}(\mathbf{k}, \lambda) = \lambda \mathbf{I} + Z[1 - \mathcal{A}(\mathbf{k})] \mathbf{W}_e(\lambda) - \mathbf{E}_e(\lambda) \quad (12.111)$$

and $\mathcal{A}(\mathbf{k})$ is defined by Eq. (12.102). We differentiate

$$\mathbf{F}(\mathbf{k}, \lambda) \mathbf{F}(\mathbf{k}, \lambda)^{-1} = \mathbf{I} \quad (12.112)$$

with respect to the components of $\mathbf{k}$, let $\mathbf{k} \rightarrow \mathbf{0}$, and note that $-\nabla_{\mathbf{k}}^2 \mathcal{A}(\mathbf{k})|_{\mathbf{k}=\mathbf{0}} = 1$, to deduce that

$$\langle \tilde{\mathbf{R}}^2(\lambda) \rangle = Z[\lambda \mathbf{I} - \mathbf{E}_e(\lambda)]^{-1} \mathbf{W}_e(\lambda) [\lambda \mathbf{I} - \mathbf{E}_e(\lambda)]^{-1} \mathbf{u}. \quad (12.113)$$

As discussed in Chapter 10, the dominant large- t behavior of $\langle \mathbf{R}^2(t) \rangle$ is deduced from the dominant small- λ behavior of $\langle \tilde{\mathbf{R}}^2(\lambda) \rangle$, although the latter must be carefully calculated since $\mathbf{E}_e(\lambda \rightarrow 0)$ is a singular matrix.

In analyzing $P(t)$, it is convenient to introduce the *propagator matrix* $\mathbf{M}_r(t)$, defined by

$$P_r(t) = \mathbf{M}_r(t) \mathbf{u} , \quad (12.114)$$

so that

$$\tilde{\mathbf{M}}_r(\lambda) = \frac{1}{(2\pi)^d} \int_{-\pi}^{\pi} \dots \int_{-\pi}^{\pi} e^{ik \cdot r} \mathbf{F}(\mathbf{k}, \lambda)^{-1} d^d \mathbf{k} . \quad (12.115)$$

After some algebra, one may show that the Laplace transform of the propagator from the origin is given by

$$\tilde{\mathbf{M}}_0(\lambda) = \mathbf{G}(\mathbf{A}) \mathbf{A} (\lambda \mathbf{I} - \mathbf{E}_e)^{-1} = \mathbf{G}(\mathbf{A}) (\mathbf{W}_e)^{-1} , \quad (12.116)$$

where, as before, $\mathbf{A} = (\mathbf{W}_e)^{-1} (\lambda \mathbf{I} - \mathbf{E}_e)$, so that $\mathbf{A}$ is a function of λ .

12.7.2.1 Effective-Medium Approximation for Conductance Disorder

We are now ready to construct a class of the EMA for the problem. We confine our attention to two cases, conveniently described as *conductance disorder*, represented by disorder in the transition rates, and *exchange disorder*.

The disorder in the transition rates or conductances corresponds to, for example, randomness in the shapes and sizes of the elements of the transport paths. For example, as we know that in rock masses, various zones of the porous matrix are usually characterized by a distribution of the permeabilities, and the fractures by a distribution of their hydraulic conductances or apertures. Thus, we assume that the exchange matrices are the same for each site (so, $\mathbf{F}_i \equiv \mathbf{0}$) and write $\mathbf{E}_i \equiv \mathbf{E}_e$. We construct the *single-bond* EMA by only allowing one bond to have a transition matrix that differs from $\mathbf{W}_e$. If this bond joins sites zero and one, Eq. (12.100) reduces to

$$\begin{aligned} \tilde{P}_i(\lambda) - \tilde{P}_i^0(\lambda) &= \mathbf{G}_{i0}(\mathbf{A}) \Delta_{01} [\tilde{P}_0(\lambda) - \tilde{P}_1(\lambda)] \\ &\quad + \mathbf{G}_{i1}(\mathbf{A}) \Delta_{10} [\tilde{P}_1(\lambda) - \tilde{P}_0(\lambda)] . \end{aligned} \quad (12.117)$$

If we recognize that $\mathbf{G}_{01}(\mathbf{A}) = \mathbf{G}_{10}(\mathbf{A})$, and let $\mathbf{G}_{00}(\mathbf{A}) = \mathbf{G}_{11}(\mathbf{A}) = -\mathbf{G}(\mathbf{A})$, we may deduce from Eq. (12.117) an *explicit* expression for the variation in $\tilde{P}_i(\lambda)$ across the 0–1 bond:

$$\tilde{P}_0 - \tilde{P}_1 = \left\{ \mathbf{I} + \frac{2}{Z} [\mathbf{I} - \mathbf{A} \mathbf{G}(\mathbf{A})] \Delta_{01} \right\}^{-1} (\tilde{P}_0^0 - \tilde{P}_1^0) . \quad (12.118)$$

To derive Eq. (12.118), we note from symmetry that Eq. (12.99) reduces for $i = k = 0$ to

$$(\mathbf{Z} \mathbf{I} + \mathbf{A}) \mathbf{G}_{00}(\mathbf{A}) - \mathbf{Z} \mathbf{G}_{01}(\mathbf{A}) = -\mathbf{I} . \quad (12.119)$$

The EMA is constructed by requiring that

$$(\tilde{P}_0(\lambda) - \tilde{P}_1(\lambda)) = \tilde{P}_0^0(\lambda) - \tilde{P}_1^0(\lambda) , \quad (12.120)$$

where the angle brackets denote the average over the disorder in the transition matrices that contain the conductances. Hence, we arrive at a working equation

$$\left\langle \left[\mathbf{I} + \frac{2}{Z} [\mathbf{I} - \mathbf{A}\mathbf{G}(\mathbf{A})] \mathbf{\Delta} \right]^{-1} \right\rangle = \mathbf{I}, \quad (12.121)$$

where $\mathbf{G}(\mathbf{A})$ is given by Eq. (12.104), and we have, for brevity, dropped the bond index 01 from $\mathbf{\Delta}_{01}$.

12.7.2.2 Effective-Medium Approximation for Exchange Disorder

This type of disorder corresponds to an asymmetry in the exchange rates between, for example, the fractures and the matrix. In this case, we assume that the transition matrices are the same for each site (so $\mathbf{\Delta}_{ij} \equiv \mathbf{0}$) and we write $\mathbf{W}_{ij} \equiv \mathbf{W}_e$. The *single-site* EMA is constructed by allowing only one site (site 0, say) to have an exchange matrix that differs from $\mathbf{E}_e$, which reduces Eq. (12.100) to

$$\tilde{P}_i(\lambda) - \tilde{P}_i^0(\lambda) = -\mathbf{G}_{i0}(\mathbf{A})\mathbf{\Gamma}_0\tilde{P}_0^0(\lambda). \quad (12.122)$$

For $i = 0$, we deduce that

$$\tilde{P}_0(\lambda) = [\mathbf{I} - \mathbf{G}(\mathbf{A})\mathbf{\Gamma}_0]^{-1} \tilde{P}_0^0(\lambda), \quad (12.123)$$

which yields the self-consistency condition

$$\langle [\mathbf{I} - \mathbf{G}(\mathbf{A})\mathbf{\Gamma}]^{-1} \rangle = \mathbf{I}, \quad (12.124)$$

where the averaging is over the disorder in the exchange matrices, and we have, for brevity, dropped the site index zero from $\mathbf{\Gamma}_0$.

Having assembled the general effective-medium formalism, we now examine some representative predictions of the approximation. Consider the case in which there is no exchange matrix (i.e., $\mathbf{E}_e = \mathbf{0}$), but allow the transition matrices $\mathbf{W}_{ij}$ to have off-diagonal terms. Then, $\mathbf{A} = \lambda(\mathbf{W}_e)^{-1} \rightarrow \mathbf{0}$ as $\lambda \rightarrow 0$, and, indeed, $\mathbf{A}\mathbf{G}(\mathbf{A}) \rightarrow \mathbf{0}$. Note that $\mathbf{G}(\mathbf{A})$ has a finite limit as $\mathbf{A} \rightarrow \mathbf{0}$ if $d \geq 3$, and a weak divergence in lower dimensions. Thus, Eq. (12.121) reduces to a simple equation for the $\lambda \rightarrow 0$ limit of the effective transition rate matrix,

$$\left\langle \left[\left(1 - \frac{2}{Z} \right) \mathbf{I} + \frac{2}{Z} \mathbf{W}_e(0)^{-1} \mathbf{W} \right]^{-1} \right\rangle = \mathbf{I}, \quad (12.125)$$

with the average taken over the distribution of the random variable $\mathbf{W}$. In space dimension $d \geq 2$, we have N^2 simultaneous nonlinear equations to solve for the N^2 elements of the matrix $\mathbf{W}_e(0)$. However, if $d = 1$ (so that $Z = 2$), the equations greatly simplify and we obtain the prediction that

$$\mathbf{W}_e(0) = \langle \mathbf{W}^{-1} \rangle^{-1}. \quad (12.126)$$

From Eq. (12.116), the propagator from the origin is given by

$$\tilde{\mathbf{M}}_0(\lambda) = \mathbf{G} [\lambda(\mathbf{W}_e)^{-1}] (\mathbf{W}_e)^{-1}. \quad (12.127)$$

If we introduce a matrix $\mathbf{V}$ to diagonalize $(\mathbf{W}_e)^{-1}$, so that

$$(\mathbf{W}_e)^{-1} = \mathbf{V} \text{diag} \{ \omega_n^2 \} \mathbf{V}^{-1}, \quad (12.128)$$

we see that

$$\langle \tilde{\mathbf{M}}_0(\lambda) \rangle = \mathbf{V} \text{diag} \{ \omega_n^2 g(\lambda \omega_n^2) \} \mathbf{V}^{-1}. \quad (12.129)$$

In one dimension, $\mathbf{W}_e(0)$ is predicted explicitly by Eq. (12.126), while $g(a) \sim 1/(2\sqrt{a})$ as $a \rightarrow 0$. Thus, we predict that

$$\langle \tilde{\mathbf{M}}_0(\lambda) \rangle = \frac{1}{2\sqrt{\lambda}} \mathbf{V} \text{diag} \{ \omega_n \} \mathbf{V}^{-1}. \quad (12.130)$$

It can be shown that this is an *exact* result.

We may derive some results for a network of arbitrary dimension d with two families of transport paths as in fractured porous media, and diagonal exchange matrices in order to have

$$\mathbf{W} = \begin{pmatrix} u & 0 \\ 0 & v \end{pmatrix}, \quad \mathbf{W}^0 = \begin{pmatrix} u^0 & 0 \\ 0 & v^0 \end{pmatrix}, \quad (12.131)$$

$$\mathbf{E} = \begin{pmatrix} -\mu & v \\ \mu & -v \end{pmatrix}, \quad \mathbf{E}^0 = \begin{pmatrix} -\mu^0 & v^0 \\ \mu^0 & -v^0 \end{pmatrix}, \quad (12.132)$$

$$\mathbf{A} = \begin{pmatrix} u^{-1}(\lambda + \mu) & -u^{-1}v \\ -v^{-1}\mu & v^{-1}(\lambda + v) \end{pmatrix}. \quad (12.133)$$

For brevity, we write

$$\kappa = \frac{\mu}{u} + \frac{v}{v} \quad \text{and} \quad \kappa^0 = \frac{\mu^0}{u^0} + \frac{v^0}{v^0}. \quad (12.134)$$

The eigenvalues of $\mathbf{A}$ are the solutions a_1 and a_2 of the equation

$$\left(\frac{\lambda + \mu^0}{u^0} - a \right) \left(\frac{\lambda + v^0}{v^0} - a \right) - \left(\frac{\mu^0}{u^0} \right) \left(\frac{v^0}{v^0} \right) = 0 \quad (12.135)$$

and so, as $\lambda \rightarrow 0$, we have

$$a_1 \rightarrow 0 \quad \text{and} \quad a_2 \rightarrow \kappa^0. \quad (12.136)$$

The corresponding eigenvectors for $\lambda = 0$ are $\begin{pmatrix} v^0 \\ \mu^0 \end{pmatrix}$ and $\begin{pmatrix} -v^0 \\ u^0 \end{pmatrix}$, and so the diagonalizing matrices are

$$\mathbf{H} = \begin{pmatrix} v^0 & -v^0 \\ \mu^0 & u^0 \end{pmatrix} \quad \text{and} \quad \mathbf{H}^{-1} = \frac{1}{\kappa^0 \mu^0 v^0} \begin{pmatrix} u^0 & v^0 \\ -\mu^0 & v^0 \end{pmatrix}. \quad (12.137)$$

Although $\mathbf{G}(\mathbf{A}) = \mathbf{H} \text{diag} \{ g(a_n) \} \mathbf{H}^{-1}$, it is not convenient to calculate $\mathbf{G}(\mathbf{A})$ this way because $g(a_1)$ diverges as $\lambda \rightarrow 0$ for $d = 1$ and two. We note, however, that as

$\mathbf{A}\mathbf{G}(\mathbf{A}) = \mathbf{G}(\mathbf{A})\mathbf{A} = \mathbf{H}\text{diag}\{a_n g(a_n)\}\mathbf{H}^{-1}$, and $ag(a) \rightarrow 0$ as $\lambda \rightarrow 0$, in the limit $\lambda \rightarrow 0$, we obtain

$$\mathbf{A}\mathbf{G}(\mathbf{A}) = \mathbf{G}(\mathbf{A})\mathbf{A} = \frac{g(\kappa^0)}{u^0 v^0} \begin{pmatrix} \mu^0 v^0 & -v^0 v^0 \\ -\mu^0 u^0 & v^0 u^0 \end{pmatrix}. \quad (12.138)$$

Similarly, in the limit $\lambda \rightarrow 0$,

$$\mathbf{G}(\mathbf{A})^{-1} = \frac{1}{g(\kappa^0)\kappa^0 u^0 v^0} \begin{pmatrix} \mu^0 v^0 & -v^0 v^0 \\ -\mu^0 u^0 & v^0 u^0 \end{pmatrix}. \quad (12.139)$$

Next, consider *exchange disorder only*, so that $\mathbf{W} = \mathbf{W}_e$ and we may therefore take $u^0 = u$ and $v^0 = v$ as known constants. The analysis is simplified by noting that since

$$\mathbf{I} = (\mathbf{W}_e)^{-1}(\mathbf{E} - \mathbf{E}_e) = \mathbf{A}(\lambda \mathbf{I} - \mathbf{E}_e)^{-1}(\mathbf{E} - \mathbf{E}_e), \quad (12.140)$$

Eq. (12.124) is equivalent to the assertion that

$$\left\langle \left\{ \mathbf{I} - \text{diag}\{a_n g(a_n)\} \mathbf{H}^{-1}(\lambda \mathbf{I} - \mathbf{E}_e)^{-1}(\mathbf{E} - \mathbf{E}_e)\mathbf{H} \right\}^{-1} \right\rangle = \mathbf{I}. \quad (12.141)$$

Although $(\lambda \mathbf{I} - \mathbf{E}_e)^{-1}$ diverges as $\lambda \rightarrow 0$,

$$(\lambda \mathbf{I} - \mathbf{E}_e)^{-1}(\mathbf{E} - \mathbf{E}_e) = \frac{1}{\mu^0 + v^0 + \lambda} \begin{pmatrix} -\mu + \mu^0 & v - v^0 \\ \mu - \mu^0 & -v + v^0 \end{pmatrix} \quad (12.142)$$

has a finite small- λ limit. After some algebra, the matrix self-consistency condition (12.141) reduces to four scalar equations:

$$\left\langle \frac{1}{D^0} \left(1 - \frac{\mu^0 - \mu}{u^0} \right) \right\rangle = \left\langle \frac{1}{D^0} \left(1 - \frac{v^0 - v}{v^0} \right) \right\rangle = 1 \quad (12.143)$$

and

$$\left\langle \frac{\mu^0 - \mu}{D^0 u^0} \right\rangle = \left\langle \frac{v^0 - v}{D^0 v^0} \right\rangle = 0, \quad (12.144)$$

with $D^0 = 1 + g(\kappa^0)(\kappa - \kappa^0)$. Although it may appear that the problem is over-determined since

$$\left\langle \frac{1 + g(\kappa^0)(\kappa - \kappa^0)}{D^0} \right\rangle = 1, \quad (12.145)$$

it is straightforward to prove that if the pair of conditions (12.144) hold, then $\langle 1/D^0 \rangle = 1$ and Eq. (12.143) are also satisfied. It follows that

$$\mu^0 = \left\langle \frac{\mu}{1 + g(\kappa^0)(\kappa - \kappa^0)} \right\rangle \quad (12.146)$$

and

$$\nu^0 = \left\langle \frac{\nu}{1 + g(\kappa^0)(\kappa - \kappa^0)} \right\rangle, \quad (12.147)$$

where κ^0 is determined from

$$\left\langle \frac{1}{1 + g(\kappa^0)(\kappa - \kappa^0)} \right\rangle = 1. \quad (12.148)$$

Although we have not exhibited it explicitly here, we emphasize that the last three equations apply only in the limit $\lambda \rightarrow 0$ (i.e., the steady-state limit). The general case of $\lambda > 0$ – the transient problem – is more complicated, but can be studied by numerical simulation.

To illustrate the predictions of the EMA, consider the case of *binary disorder* where (μ, ν) takes the values (μ^*, ν^*) and $(0, 0)$ with probabilities p and $1 - p$ respectively, and for brevity, write $\kappa^* = \mu^*/u^0 + \nu^*/\nu^0$. Equation (12.148) becomes

$$(1 - p) = \left(1 - \frac{\kappa^0}{\kappa^*}\right) [1 - g(\kappa^0)\kappa^0]. \quad (12.149)$$

This leads, in one dimension, to a quadratic equation for κ^0 . However, in higher dimensions, the scalar lattice Green function g is not an elementary function and the equation must be solved numerically. The more general case in which (μ, ν) takes the values (μ_1, ν_1) and (μ_2, ν_2) with probabilities p and $1 - p$, respectively, where $\nu_1 \ll \mu_1$ and $\nu_2 \ll \mu_2$, is of practical interest as it correspond to transport in a porous catalyst with micropores and macropores, or a natural porous medium with tight pores and large fractures.

Next, consider two-path porous media – those with pores and fractures – with conductance disorder only, which represent a more subtle case. If we attempt to write $\mu^0 = \mu = \text{constant}$, we find that the self-consistency condition (12.121) reduces in the $\lambda = 0$ limit to *three* independent scalar equations for the *two* unknown functions $u^0(0)$ and $\nu^0(0)$ that, in general, admit no solution. This suggests that for conductance disorder, the uniform system with the macroscopic transport properties of the disordered porous media must either possess off-diagonal terms in the matrix $\mathbf{W}_e(\lambda)$, or have an exchange matrix $\mathbf{E}_e(\lambda)$ that differs from that of *each realization* of the media. In other words, matching a disordered medium with several distinct families of transport paths to a uniform one induces not only memory, but also *additional couplings* that are absent from the original medium. This explain why simply coupled diffusion equations that are used in the continuum multiporosity models described above often are poor models for transport in fractured porous media. Such a subtle feature is perhaps the most important insight provided by the network formulation of the problem.

Even the $d = 1$ limit of this problem with $N \geq 2$ is nontrivial and far more complex than the $N = 1$ case. For example, whereas percolation-like disorder divides a 1D system into finite segments and prohibits macroscopic transport, for $N \geq 2$, the system can still have global transport even if m ($m < N$) paths have been disrupted by percolation disorder. A more interesting case arises if transport in one

family of paths is much faster than in the other(s). For example, flow and transport in a fracture network are much faster than those in the matrix. As we learned in Chapter 6, in natural porous media, it is often true that the fracture network has a fractal structure. Moreover, at the largest length scales (of the order of a kilometer or more), the fracture network may have the structure of the sample-spanning percolation cluster at the percolation threshold (see Chapters 6 and 8), while the pore network is well-connected. In this case, one may expect a variety of interesting results to emerge. In particular, the system may behave quite differently on various time scales that would depend on how the transport process is partitioned between the two networks, and whether the two networks are macroscopically connected. Using the exact formulation and the EMA described here, one may study such issues.

Experimental evidence for the formulation presented above was provided by Mattisson *et al.* (1997), who measured the permeability and resistivity of sintered porous media with disordered fractures over a wide range of matrix porosity. Cáceras (2004) carried out a thorough analysis of the Hughes–Sahimi model and presented a number of extensions.

13

Miscible Displacements

Introduction

In Chapters 11 and 12, we described and studied dispersion processes in flow through porous media, fractures and fracture networks, and fractured porous media. Dispersion involves two miscible fluids that have the same density and viscosity and one fluid phase. In the present chapter, we describe and study displacement of one fluid by another miscible fluid with viscosity, and possibly density, different from those of the fluid to be displaced.

If we inject a fluid into a porous media that is saturated with a second fluid, and if the two fluids mix in all proportions and their mixture remain a single phase, the two fluids are said to be *first-contact miscible*. Intermediate-molecular weight hydrocarbons, for example, propane and butane, have such a property. In other situations, the injected and in-place fluids may form two different phases, that is, they are not first-contact miscible. However, mass transfer between the two phases and repeated contact between them can achieve miscibility. This is usually called *multiple contact* or *dynamic miscibility*. In the petroleum industry, the miscible injection fluids that achieve either first-contact or dynamic miscibility are usually called miscible solvents. In the present chapter, we restrict ourselves to first-contact miscible problems, as modeling multiple-contact miscibility involves thermodynamic phase equilibria calculations that are beyond the scope of this book.

Why are miscible displacements important? On average, two-thirds of the original oil in any oil reservoir remains unrecovered, even after water injection into the reservoir. The same is true if a low-pressure gas, which is largely immiscible with the oil, is injected into the reservoir. After the primary recovery ends, the remaining oil becomes trapped in the reservoir due to the capillary forces and the interfacial tension, and remains entrapped regardless of how much water or low-pressure gas is injected into the reservoir. It forms either a discontinuous phase in the swept zone, or a continuous phase in the unswept zone of the reservoir. One efficient way of enhancing the recovery of oil is by injecting into a reservoir a fluid that is miscible with the oil.

In principle, the injected fluid should significantly reduce the capillary and interfacial forces. If that happens, then one may recover a large fraction of the trapped oil. For example, if one injects a gas (or a gas mixture) into an oil reservoir and if,

at the reservoir's temperature and pressure, the gas is in a critical or near critical state, it will mix in large proportions (if not completely) with the oil in place. Under such conditions, one will have an efficient miscible displacement process. For example, the critical temperature of CO₂ is only about 31 °C and, therefore, it can be an ideal agent for miscible displacement of oil. In fact, field-scale CO₂ injection has been successfully carried out in the United States.

A typical miscible injection, or flooding, is carried out by injecting a limited volume or slug of the solvent into the reservoir. The solvent slug may be displaced miscibly by an appropriate drive fluid (miscible slug process), or immiscibly by, for example, water. The latter process leaves a residual solvent saturation in the reservoir and causes dilution and fingering of the drive fluid in the solvent, and fingering of the solvent in the oil, all of which reduce the overall effectiveness of the process.

Such miscible displacements have received considerable attention since the early 1950s. Over 100 studies were undertaken in the 1950s and early 1960s to investigate the feasibility and economics of miscible displacement processes, particularly gas injection, as an effective tool for enhanced oil recovery (EOR). The early studies indicated that natural gas, flue gas, nitrogen at high pressure, and enriched hydrocarbon gases achieve miscibility with the reservoir oil. A slug of fluids containing oil, water, surfactants, and co-solvents (for example, alcohols) in various compositions – known as micellar polymer solutions – have also been found to be efficient miscible displacement agents, although they are not used commonly, due to their cost.

The best advantage of N₂ flooding is perhaps the fact that it can potentially be used anywhere in the world if it can be cheaply extracted from the air, whereas other injection fluids are either not available or the cost of their delivery to the oil reservoir is prohibitive. On the other hand, since light hydrocarbons contain considerable combustion energies, their use as EOR agents is generally limited to remote locations, which makes the cost of delivery of the oil to the market too high. In the United States, the main emphasis has been on miscible CO₂ flooding since it offers two main advantages over other gases. One is that CO₂ requires a relatively low operating pressure to attain miscibility with reservoir fluids. The second advantage is that CO₂ is relatively inexpensive as it has no value as a fuel, and is available in large amounts from natural deposits or as a waste product of industrial processes. In addition, injection of CO₂ into an oil reservoir is increasingly being viewed as a viable method of its sequestration and lessening its impact on the climate and the Green House effect.

However, even if the gas as the miscible displacement agent is economically available, its use is still not without problems. Gases are normally less viscous than typical crude oil. The viscosity contrast between the oil and the injected gas, together with the phenomenon of *gravity segregation* (see below), render the miscible displacement much less efficient than desired. In addition, in most miscible displacement processes involving a gas, the required temperature and pressure for miscibility of the oil and the displacement agent are often high enough that they limit the number of prospective reservoirs. For example, medium to heavy hydro-

carbons become miscible with oil only at high temperatures and/or pressures (see, for example, Sahimi *et al.*, 1985; Sahimi and Taylor, 1991). Another negative aspect of a miscible displacement process is its cost. It may happen that a miscible displacement is more efficient in terms of the amount of the recovered oil than an immiscible injection, but the total cost of the miscible displacement (including the cost of transporting the displacing gas to the oil field from other locations) is so high that it renders it economically unattractive. Moreover, flue gas has only limited application as an agent of a miscible displacement in deep and high pressure reservoirs. For these reasons, the EOR processes based on gas injection have not been as common as immiscible displacement processes, such as, water flooding, although they are used relatively extensively.

13.1

Factors Affecting the Efficiency of Miscible Displacements

Miscible injection as an EOR process is influenced by several factors. Although such factors are well-known, there is still some disagreement on the extent of the influence of each individual factor. For example, while *laboratory* experiments indicate that the viscosity contrast between the oil and the displacing gas has a strong effect on the efficiency of the miscible displacements, the same may not be true at the *field scale* that is dominated by large-scale heterogeneities, for example, the spatial distributions of the permeability and porosity and the presence of fractures and/or faults. In what follows, we describe the effect of some of the most important factors on the efficiency of a miscible displacement.

13.1.1

Mobility and Mobility Ratio

The mobility λ_i of a fluid i is defined as the ratio of the effective permeability K_i of the porous medium, experienced by fluid i , and the fluid's viscosity μ_i , $\lambda_i = K_i/\mu_i$. When one fluid displaces another, the mobility ratio M is defined as the ratio of the mobilities of the displacing and displaced fluids, and is one of the most important influencing factors in any displacement process. Normally, M is not constant because mixing of the displacing and displaced fluids changes the effective viscosities of the mixed region. In addition, the viscosity of the mixed zone also depends on the concentrations of the displacing and displaced fluids. In many cases, the viscosity μ_m of the mixed zone is estimated from the following empirical law due to Koval (1963)

$$\mu_m = \left(\frac{C_s}{\mu_s^{\frac{1}{4}}} + \frac{1 - C_s}{\mu_o^{\frac{1}{4}}} \right)^{-4}, \quad (13.1)$$

where C_s is the solvent concentration, and μ_s and μ_o are the viscosities of the solvent and oil, respectively. Sometimes, instead of C_s , the solvent volume fraction is

used in Eq. (13.1). If, in addition to the solvent, another fluid, such as water, is also injected into the porous medium, as is often done in order to reduce the mobility of the solvent, then defining an effective value of M can become problematic.

The mobility ratio when a solvent displaces oil at irreducible water saturation – the saturation at which the water phase becomes disconnected in the pore space (see Chapter 14) – with negligible mixing of the solvent and oil, is simply the ratio of oil and solvent viscosities, $M = \mu_o/\mu_s$, assuming that the porous medium is homogeneous, though microscopically disordered, so that the permeabilities K_o and K_s experienced by the two fluids are essentially equal. M is always greater than unity and if $M > 1$, the mobility ratio is said to be *unfavorable* since it may lead to formation of fingers that reduce the efficiency of a miscible displacement (see below). Defining an effective mobility ratio is more complex and uncertain when mobile water is present.

In many miscible displacement processes, one must deal with more than one displacing front. For example, tertiary oil recovery (which is carried out when, for example, water flooding is no longer effective) usually involves more than one displacing fronts. The problem of defining an effective M for such situations is even more complex, and no completely satisfactory method has been developed yet for addressing the problem. In such processes, motion of any given front between the two fluids is affected not only by the mobility ratio across that front (i.e., the mobility ratio of the two fluids on the two sides of the front), but also by the mobilities of the other regions behind and ahead of the front as well as by their relative sizes.

13.1.2

Diffusion and Dispersion

Under certain conditions, mixing of the displacing and displaced fluids by diffusion can be important. For example, during both secondary and tertiary displacement of a reservoir's oil by CO_2 , the development of multiple-contact miscibility strongly controls the ultimate recovery efficiency. At the microscopic scale, molecular diffusion is the mechanism by which molecular mixing of CO_2 and oil occurs. It is at this scale that the usual assumption of rapid local equilibrium is made, and is used for numerical simulation of CO_2 flooding. Similarly, during flooding of rich gases that contain hydrocarbons of intermediate molecular weights, miscibility with the in-place oil is developed by a multiple-contact condensing mechanism, during which diffusive mass transfer plays an important role. Moreover, a significant oil saturation may exist in the dead-end pores, or be trapped by water films in a water-wet porous medium during a miscible displacement. Such isolated oil remains largely unrecoverable unless the fluid injected into the porous medium can efficiently traverse the surrounding water barriers to contact and swell the trapped oil. The injected fluid also penetrates the oil by molecular diffusion that, in turn, inhibits viscous fingering (since diffusion helps the fingers join together), delays premature breakthrough and, therefore, increases the oil production rate.

If two miscible fluids with an initially sharp front separating them are put in contact, their subsequent mixing caused by molecular diffusion is described by the

diffusion equation:

$$\frac{\partial G_i}{\partial t} = -D_{io}A \frac{\partial C_i}{\partial x}, \quad (13.2)$$

where G_i is the amount (in moles, for example) of fluid i that has diffused across the front at time t , D_{io} is the effective diffusion coefficient of fluid i in the porous medium, A is the cross-sectional area for diffusion, and C_i is the molar concentration of i at position x at time t . The diffusion coefficient D_{io} depends, in principle, on the mixture's composition. However, in reservoir simulations, an average diffusion coefficient at 50% solvent concentration usually yields adequate representation of the diffusive mixing. Many experimental methods have been developed for measurement of the effective diffusion coefficient D_{io} involving oil in porous media (Gavalas *et al.*, 1968; Renner, 1988; Nguyen and Farouq-Ali, 1995). Experimental data for the effective diffusivity are still necessary because as the discussions in Chapters 9 and 10 indicated, despite several decades of research, no accurate theoretical method for estimating the effective diffusivities of mixtures in porous media is yet available. Unfortunately, even measurements are generally difficult and very time consuming. Most conventional methods require composition analysis that is tedious and expensive (Moulu, 1989). Simpler methods of measuring the effective diffusion coefficients for a gas-oil mixtures have also been proposed (Riazi, 1996; Zhang *et al.*, 2000).

As described in Chapter 11, dispersion is mixing of two miscible fluids flowing in a system, for example, a porous medium. Therefore, unlike diffusion, the velocity field of the fluids plays an important role in this type of mixing process. Similar to diffusion, mixing by dispersion can decrease the viscosity and density contrasts between the displacing and the displaced fluids, which in most cases is very useful to the displacement process.

As the two major mechanisms of mixing by dispersion are small- and large-scale variations of fluid velocities (or, equivalently, the permeabilities) and molecular diffusion, they both help mixing of the two miscible fluids. If, for example, a first-contact miscible solvent is injected into a linear packed-bed column to displace oil, the effluent concentration profile of the solvent will have an S-shape, which is the result of mixing the solvent and oil in the packed-bed. Because of this, a transition zone of solvent/oil mixtures separates a zone of 100% solvent from one that is pure oil. This mixing, which is in the direction of the macroscopic flow, is the longitudinal dispersion that we studied in Chapter 11.

As Chapter 11 taught us, transverse dispersion also occurs in the direction(s) perpendicular to the direction of macroscopic flow, which occurs when, for example, a solvent is injected into a stratified porous medium that consists of layers of different permeabilities parallel to the macroscopic flow. In this case, the solvent in the more permeable layer moves faster and mixes with the oil, not only by longitudinal dispersion in the direction of flow, but also by transverse dispersion perpendicular to the direction of flow in the less permeable layers.

Since the longitudinal dispersion coefficient D_L is usually larger than the transverse dispersion coefficient D_T by a factor that, depending on the morphology

of the porous medium, ranges anywhere from 5 to 24, the large difference between the two implies that more mixing occurs in the direction of the macroscopic flow. Since pore space heterogeneities strongly affect D_L and D_T (see Chapters 11 and 12), the implication is that the heterogeneities also affect miscible displacements. Unlike the effective diffusivities, the dispersion coefficients D_L and D_T depend on the mean flow velocity. As discussed in Chapter 11, the relation between the dispersion coefficients and the mean flow velocity depends on the value of the Péclet number Pe , defined as $Pe = \mathcal{L}V/D_m$, where $\mathcal{L}$ is a characteristic length scale of the porous medium, and D_m is the molecular diffusivity. The relation between the dispersion coefficients and Pe is, in general, nonlinear (see Chapter 11), but it is often assumed in numerical simulations that $D_L = \alpha_L V$ and $D_T = \alpha_T V$, where α_L and α_T are the corresponding dispersivities. Roughly speaking, if the length scale over which the measurements are done is larger than the dispersivities, then the convective-diffusion (CD) equation may be used to describe the dispersion process and, thus, miscible displacements.

Use of constant dispersivities permits a phenomenological description of transport through porous media. A fundamental understanding of the phenomena is obtained, however, only if the dispersivities are related to the basic properties of porous media, for example, their porosity, hydraulic conductivity and/or pore size distribution. As described in Chapter 11, to develop such relations, two approaches have been developed in the past. One approach constructs empirical correlations between the measured dispersivities and important morphological parameters of the porous medium. For example, Harleman and Rumer (1963) found, based on the analysis of their experimental data, that the longitudinal dispersivity is proportional to the square root of the mean hydraulic conductivity of their porous medium. Attempts to verify their proposed relationship under field conditions (Taylor *et al.*, 1987) using the results of tracer tests were not, however, successful. The second approach is theoretical, using various models of heterogeneous porous media that were described in Chapters 11 and 12.

It is important to recognize that values of the dispersion coefficients measured in the laboratory are usually *smaller* than the corresponding field-scale values. This presumably explains why the empirical relations between the dispersivities and the hydraulic conductivity (obtained based on laboratory experiments) fail under field conditions. Thus, one must devise an appropriate method for scale-up of the dispersion coefficients from the laboratory to the field scales (Johns *et al.*, 2000). Mixing by dispersion in a field-scale porous medium is likely to be much greater than that in a laboratory-scale sample of the same porous medium since large-scale variations in the permeability and porosity of the field-scale porous medium strongly increase the mixing process at that scale.

Mobility ratio and gravity also affect dispersion. If $M > 1$, viscous instability develops (see below), in which case the displacement is no longer a simple process. If, however, $M < 1$, the usual dispersion mechanisms described in Chapter 11 are operative. Moreover, since no instability develops for $M < 1$, the effect of pore space heterogeneities is also suppressed. On the other hand, if a less dense fluid

displaces a denser one, the density difference suppresses the effect of dispersive mixing.

In some situations, longitudinal dispersion affects a miscible displacement more strongly than the transverse dispersion, and vice versa. For example, if large fingers of the displacing fluid develop, which is often the case when $M > 1$, or when there are large-scale variations in the permeabilities, there would be a large surface area on the sides of the fingers that allows for significant transverse dispersion to occur. This can help join the fingers, stabilize the displacement, and increase its efficiency. By contrast, longitudinal dispersion can only take place at the tip of the fingers and, therefore, its effect is much weaker than that of transverse dispersion. For this reason, models that ignore transverse dispersion are usually not adequate for describing a miscible displacement.

13.1.3

Anisotropy of Porous Media

As discussed in Chapter 11, dispersion processes are sensitive to the distribution of the heterogeneities of a porous medium, including its stratification that, in turn, affects the performance of miscible displacements. This is particularly true when the displacing agent is a gas. It should be clear that a displacing gas preferentially chooses the strata with higher permeabilities. As a result, large amounts of the fluid (oil) to be displaced may be left behind in the strata with low permeabilities. If we attempt to displace the by-passed fluid (oil) by injecting larger amounts of gas into the low permeability strata, some of the gas will inevitably enter the high permeability strata and do essentially nothing as such strata have already been swept by the previously-injected gas and, therefore, additional gas injection would not be very efficient. The effect of stratification is even stronger when the mobility ratio $M > 1$.

Another phenomenon that affects miscible displacements in stratified media is the crossflow of displacing and displaced fluids between the strata. Depending on the direction of the displacement process, crossflows can help or hinder the efficiency of the process. We will come back to this shortly.

13.2

The Phenomenon of Fingering

If the injected fluid and the fluid that it displaces in a porous medium are first-contact miscible, and if the mobility ratio $M < 1$, then the displacement process is very simple and efficient. The displaced fluid moves ahead of the displacing fluid, and the displacement front is stable and, aside from small perturbations, relatively smooth. There is also a mixed zone between regions of pure displacing and displaced fluids. However, in practice, a miscible displacement process is not so simple. Since, typically $M > 1$, the front is unstable and many fingers of the mixture of the displacing and displaced fluid develop, leaving behind large amounts

of oil or any other fluid to be displaced. The formation of the fingers which have very irregular shapes strongly reduces the efficiency of the miscible displacements. Figure 13.1 presents the effect of the mobility ratio M on the formation and shape of the fingers. The experiments were carried out in a quarter of the *five-spot geometry* that was made of consolidated sand, that is, a geometry in which the injection point (or well in a field application) is at the center of the system and four production points (wells) are at the four corners of the system. The porous medium in these experiments was essentially two dimensional (2D). As Figure 13.1 indicates, as M increases, the amount of swept oil decreases, and thin and long fingers of the displacing fluid are formed.

Such a phenomenon is usually referred to as *viscous fingering*. The terminology is an accurate representation of the phenomenon in macroscopically-homogeneous porous media, for which the mobility ratio reduces to the ratio of the viscosities of the displaced and displacing fluids. Under field conditions, fingering may be dominated by the distribution of the heterogeneities of the porous formation and, therefore, one should simply refer to the phenomenon as *fingering*.

Why do viscous fingers form? Collins (1961) gives a simple but very clear description of the physics of the phenomenon. Suppose that a porous medium is saturated with oil, and a displacing or solvent fluid is injected into the medium to displace it. Assume also that dispersion is negligible. The displacing fluid displaces the oil linearly, that is, if the porous medium is homogeneous, the front will remain a flat plane throughout the process. Suppose now that the displacing fluid encounters a small region of higher permeability. Then, the front will travel faster in that region and produce a bump that is a distance ϵ ahead of the rest of the front. If K_e , ϕ , and ΔP are, respectively, the effective permeability, porosity, and pressure difference along the porous medium, then, using Darcy's law, we write

$$\frac{dx_f}{dt} = \frac{K_e \Delta P}{\mu_s \phi [ML + (1 - M)x_f]}, \quad (13.3)$$

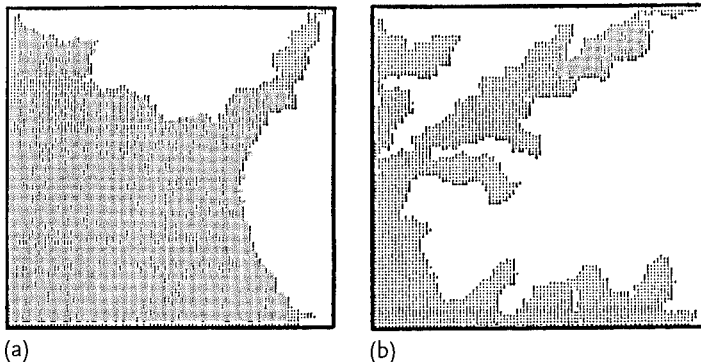


Figure 13.1 Patterns of viscous fingers for $M = 1$ (a) and $M = 10$ (b). The dark area represents the displacing fluid.

when x_f is the position of the front, L is the medium's length, and $M = \mu_0/\mu_s$. Similarly, we may write

$$\frac{d(x_f + \epsilon)}{dt} = \frac{K_e \Delta P}{\mu_s \phi [ML + (1 - M)(x_f + \epsilon)]}, \quad (13.4)$$

and, therefore

$$\frac{d\epsilon}{dt} = \frac{-K_e \Delta P(1 - M)}{\mu_s \phi [ML + (1 - M)x_f]^2} \epsilon = c\epsilon, \quad (13.5)$$

if $\epsilon \ll x_f$. Hence, $\epsilon = \exp(ct)$. However, $c > 0$ if $M > 1$, which means that ϵ grows exponentially with the time t , and a long thin finger is formed. However, the finger is not formed, or if it does, it dies out if $M < 1$. Although the analysis is overly simplified, it does illustrate the effect of M on the formation of viscous fingers. We should remark that finger formation can also occur in immiscible displacements that will be described in Chapter 14.

Due to its significance, fingering has been studied for a long time. Several older reviews provide detailed discussions of the subject. Among these are those of Wooding and Morel-Seytoux (1976), who reviewed viscous fingering in a porous medium, Bensimon *et al.* (1986) and Saffman (1986), who discussed immiscible viscous fingering only in a Hele-Shaw cell, and Homsy (1987), who considered the problems in both Hele-Shaw cells and porous media. A Hele-Shaw cell (Hele-Shaw, 1898)¹⁾, is an essentially 2D system confined between two flat plates of length L that are separated by a small distance b . Flow in the cell can be rectilinear, that is, the fluid is injected into the system at one face of it and produced at the opposite face, or it can be radial in which the fluid is injected into the system at its center. Viscous fingering also belongs to the general class of pattern-selection problems such as crystal growth phenomena.

As long as mixing due to dispersion is absent, the analogy between formation of fingers in Hele-Shaw cells and a 2D porous medium is valid, which is why the study of miscible displacements in Hele-Shaw cells was popular. If dispersion is present, the analogy breaks down because transverse dispersion is always present in a porous medium, whereas a Hele-Shaw cell with its thin gap between the two plates cannot support significant transverse dispersion in the third direction. There have also been many experimental studies of fingering phenomena in both miscible and immiscible displacements. They include those of Blackwell *et al.* (1959), Benham and Olson (1963), Slobod and Thomas (1963), Greenkorn *et al.* (1965), Kyle and Perrine (1965), Perkins *et al.* (1965), Mahaffey *et al.* (1966), Perkins and Johnston (1969), and more recently, those of Paterson (1981, 1983, 1985), Paterson *et al.* (1982), Chen and Wilkinson (1985), Lenormand *et al.* (1988), and Bacri *et al.* (1991). Most of the experiments were also accompanied by a linear stability analysis (see below).

1) Henry Selby Hele-Shaw (1854–1941), holder of the Harrison Chair of Engineering at Liverpool University College, was a mechanical engineer. In addition to his work on fluid flow in thin cells, he invented the variable-pitch propeller that contributed to Britain's success in the Battle of Britain in 1941.

Before we go on with the description of miscible displacements, we should mention that Hill, S. (1952) appears to be the first who published experimental and simple analytical results on viscous fingering and its stability. Later, Saffman and Taylor (1958) and Chouke *et al.* (1959) carried out the first rigorous analysis of the problem (although they considered immiscible viscous fingers studied in Chapter 14). Homsy (1987) suggested that one must call this phenomenon the *Hill instability* problem instead of the popular *Saffman–Taylor instability*. We take a middle-of-the-road approach and refer to it as the *Hill–Saffman–Taylor instability*.

13.3

Factors Affecting Fingering

Since various gases, and in particular CO₂, have been used as the displacing agent in miscible displacements, it is worthwhile to consider the factors that influence the fingering phenomenon, that is, when a gas is injected into a porous medium. Generally speaking, the fingers' patterns are influenced by the fluids' characteristics, including the possible nonmonotonicity in their viscosity profile (Manickam and Homsy, 1993), and the heterogeneity of the porous medium manifested by the variance and correlation structure of the permeability field (Araktingi and Orr, 1988; Waggoner *et al.*, 1992; Sorbie *et al.*, 1994). Fingering is mitigated by small-scale dispersion (at the pore and laboratory scales) and by large-scale variations in the permeability and channeling.

13.3.1

Displacement Rate

Compared to immiscible displacements, miscible displacements are much less sensitive to the displacement rate. Fingers that emerge during miscible displacements are only mildly sensitive to the displacement rate. This is due to the fundamental role that dispersion-driven mixing plays that help the smaller fingers to merge. The mild sensitivity of the miscible fingers to the displacement rate has practical implications. To control the stability of an immiscible displacement, it is often enough to control the displacement rate. This is clearly not the case with miscible displacements. While it is possible to obtain a stable displacement by using a rate less than a critical rate, in most practical situations, this would imply using a rate that would not be economical.

13.3.2

Heterogeneity Characteristics

As pointed out earlier, one factor that plays a fundamental role in finger formation is the heterogeneity of the porous medium. Once a finger begins to grow, its subsequent growth is closely linked to its interaction with the heterogeneity and, in particular, to the spatial variations of the permeability. Permeability variations

have been found to play an important role in finger initiation and growth (Stalkup, 1984; Moissis *et al.*, 1987). The spatial variation of the permeability is usually described by two parameters. One is the index of variations, already introduced in Section 5.2.3. Also called the coefficient of permeability variation H_K , it may be defined by a variety of ways, for example,

$$H_K = \frac{\sigma_K}{K_m}, \quad (13.6)$$

where σ_K is the standard deviations of the permeability distribution, and K_m is its mean (or expected value). The second characteristic quantity is a permeability correlation length ξ_K , which is the length scale over which the permeabilities are spatially correlated.

Moissis *et al.* (1987) found that the permeability distribution near the inflow end of their 2D model determines the number of the fingers, their initial locations, and their relative growth rates. The locations of the fingers are controlled by the maxima in the permeability distribution near the inflow end. The initial number of the fingers depends strongly on the correlation length of the permeability distribution. Highly-correlated porous media have fewer maxima in their permeability distribution and, consequently, fewer fingers form in such porous media. As mentioned above, some of the initial fingers grow faster than the rest and eventually dominate the displacement. Due to the *shielding effect*, the long fingers suppress the growth of small fingers (see also below). The smaller fingers may merge later on with the larger ones, leaving unswept areas that may be fairly extensive. This process results in a number of large fingers that grow quite independently of each other, at least until the breakthrough, and are referred to as the *active fingers*.

The number of the active fingers is a decreasing function of the permeability correlation length ξ_K since, as discussed above, the initial number of the fingers is smaller for larger ξ_K . Moreover, the effect of downstream permeability distribution (see below) is more significant for large ξ_K , resulting in more merger of the fingers, which reduces their number. For a given value of H_K , the grow rate of the fingers increases with ξ_K up to a limiting value. For large values of ξ_K , the permeability variation tends to generate fingers of large wavelengths that grow relatively slowly. However, once such fingers grow beyond the zone in which their growth is approximately described by the linear stability analysis (see below), it is easier for the displacing gas to develop flow channels that accelerate the subsequent growth rate of the fingers.

The number of active fingers is also a decreasing function of H_K . As H_K increases, the difference in permeability between high- and low-permeability regions increases. In the initial stages of the displacement, the difference favors the growth of the longest fingers which tend to grow in the high-permeability regions. Thus, for highly heterogeneous porous media, the longest fingers grow much faster than the rest and dominate the displacement relatively early, shielding the smaller fingers. Later on, during the displacement, the large difference in the permeabilities from region to region facilitates merging of the fingers. The net result of the two mechanisms is the reduction of the number of active fingers. Since almost all the

displacing fluid flows through one or at most a few fingers, breakthrough occurs early and, therefore, the sweep efficiency is poor.

The effect of downstream permeability variations on the finger formation and growth is negligible for random (uncorrelated) porous media, but it becomes increasingly more important as the correlation length ξ_K increases. This effect is more pronounced for small viscosity ratios. The cause of such behavior is traced to the scale of the permeability variations. For a finger to be significantly affected by the permeability variations, the length scale over which the variations are significant must be of the same order of magnitude as the finger's width or larger. In porous media with a correlated permeability distribution, the existence of high- and low-permeability regions causes flow channeling and, thus, affects finger growth mainly by causing additional merging of the fingers. For uncorrelated porous medium, on the other hand, the fingers' width is always larger than the length scale over which the permeability variations are significant.

13.3.3

Viscosity Ratio

As discussed earlier, one of the main characteristics of finger growth is that the large initial number of fingers is reduced by their merging and formation of a smaller number of active fingers. The effect of the viscosity ratio on such a process is important: At low viscosity ratios, there are no fingers that grow dramatically faster than the rest. Consequently, finger merging and growth suppression occur to a much lesser extent, resulting in a larger number of active fingers with growth rates that do not vary much. At high viscosity ratios, on the other hand, the active fingers start to outgrow the others earlier, and more finger merging and suppression of growth occur. The net result is a smaller number of long active fingers.

13.3.4

Dispersion

Strong longitudinal dispersion helps formation of a thick (more diffused) displacement front, hence making the fingers less susceptible to flow disturbances that are caused by small-scale heterogeneities. Moreover, since the front is more diffused, the waves that correspond to small values of the concentration (of the displacing fluid) grow faster and, therefore, the breakthrough occurs earlier. While it may appear that the fingers should grow faster when longitudinal dispersion is larger, it is actually difficult to define finger lengths or growth rates in this case. The effect of the transverse dispersion on the fingers was already described.

13.3.5

Aspect Ratio and Boundary Conditions

We define the aspect ratio $\mathcal{R}_0$ of a porous medium as the ratio of its length and width, $\mathcal{R}_0 = L_x/L_y$. If $\mathcal{R}_0$ increases (narrower porous media, somewhat like a slim

tube commonly used in laboratory studies of CO₂ injection), the initial fingers are closer to one another and, consequently, their interaction is stronger. The interactions result in suppression of the growth of the smaller fingers and their merging at the initial stages of the displacement, hence yielding, even at early times, a small number of active fingers. At very large aspect ratios, only one active finger will form.

The effect of the geometry of a porous medium on fingering phenomena has received considerable attention (Zimmerman and Homsy, 1991; Waggoner *et al.*, 1992; Sorbie *et al.*, 1994). Typically, simulations of unstable miscible displacements at field scales are carried out in geometries that have an aspect ratio of about three (Christie, 1989). Waggoner *et al.* (1992) simulated displacements at conditions of transverse (or vertical) equilibrium. Such a limit is reached when the generalized aspect ratio, $\mathcal{R} = (L_x/L_y)(K_v/K_h)^{1/2}$, is large, where K_h and K_v are the horizontal and vertical permeabilities. Sorbie *et al.* (1994) studied the sensitivity of the displacement patterns to $\mathcal{R}$ in heterogeneous reservoirs, and showed that $\mathcal{R}$ significantly affects the delineation in the parameter space of the various displacement regimes.

In another study, Yang and Yortsos (1995, 1996) provided an asymptotic description of the displacements in porous media, including formation of fingers in the limit that the parameter $\mathcal{R}$ is large or small. Large $\mathcal{R}$ corresponds to conditions of transverse equilibrium, which is reached in long and narrow isotropic reservoirs in those in which the permeability transverse to the applied pressure gradient significantly exceeds the permeability parallel to it, and in slim tubes. It is a regime in which intense transverse mixing occurs. Small $\mathcal{R}$ corresponds to the opposite regime of zero transverse mixing and is better known as the *Dykstra–Parsons approximation* (Dykstra and Parsons, 1950).

In parallel, Yang (1995) reported on the sensitivity of viscous fingering to $\mathcal{R}$ by means of high-resolution simulations. He reported that for uncorrelated, weakly heterogeneous porous media at conditions near transverse equilibria, most of the viscous fingering ultimately occurs near the lateral, no-flow boundaries of the system. More specifically, he found that narrow, single fingers originate at such boundaries and propagate faster than the fingers in the interior of the domain until a small permeability value is randomly encountered, at which point the fingers turned inwards. The intensity of this effect was found to depend on the viscosity ratio, and on the heterogeneity parameter. Yang and Yortsos (1998, 2002) showed that this effect is not a numerical artifact, but arises as a consequence of the slip boundary condition implied by the use of Darcy's law along no-flow boundaries. They found that the origin of the boundary effect is the vanishing of the transverse, but not of the streamwise, velocity component at the no-flow boundary. When $\mathcal{R}$ is small (for example, when $K_v < K_h$), transverse mixing is minimal everywhere (including the boundary), and so also is the boundary effect. By contrast, for large $\mathcal{R}$ (for example, when $K_v > K_h$), transverse mixing is intense everywhere, except at the no-flow boundary. Therefore, the growth of all the fingers, except the one at the boundary, is mitigated.

13.4

Gravity Segregation

An important factor that influences vertical sweeps in miscible displacements is the gravity. Solvents are usually less dense than either oil or brine, and drive gases, such as hydrocarbons or flue gas, are even less so. Due to the density differences, solvents and drive gases may segregate and override the other reservoir fluids that, in turn, decrease the vertical sweep in horizontal floods. On the other hand, gravity can be used as an advantage in dipping reservoirs to improve the sweep and the displacement efficiency.

To study the effect of gravity on miscible displacements, two dimensionless numbers are introduced:

$$\eta = \frac{(\rho_1 - \rho_2)}{\rho_2}, \quad (13.7)$$

$$N_G = \frac{gK_e(\rho_1 - \rho_2)}{q\mu_1}, \quad (13.8)$$

where N_G is called the *gravity number*, subscript one denotes the displaced fluid (e.g., oil), while two denotes the displacing fluid. If N_G is small, gravity is unimportant and viscous fingering dominates the flooding behavior. As N_G increases, viscous fingering can still occur, but gravity influences the growth rates of the individual fingers. If the solvent is lighter than oil, it tends to flow upwards. Hence, more solvent enters the fingers in the upper part of the porous medium, resulting in faster growth of these fingers, while the growth of the rest of the fingers is somewhat suppressed, partly due to gravity drainage and partly because of the shielding effect described earlier. This phenomenon results in early breakthrough and reduced sweep efficiency. If, however, the solvent is heavier than oil, it is conceivable that, for a range of N_G , gravity might improve the sweep efficiency by delaying the breakthrough of the fastest-growing finger.

If N_G is large enough, a *gravity tongue* is formed and grows at the top of the porous medium. Moreover, viscous fingering may occur near the tongue, while fingering in the rest of the medium will be suppressed. Under this condition, both gravity override and viscous fingering are important and affect the displacement. For very large N_G , gravity override completely dominates the displacement, suppressing any fingers that may form at early times. In this case, the gravity tongue breaks through very early, and the rate of oil recovery will be very low. Therefore, it should be clear that as N_G increases, the breakthrough time decreases, while the rate of oil recovery after the breakthrough also decreases (Moissis *et al.*, 1989).

The effect of gravity is more pronounced at high viscosity ratios because the ratio of gravitational and viscous forces is inversely proportional to the viscosity of the fluid currently present in the porous medium. This ratio is equal to N_G only at time $t = 0$. As the displacement proceeds and the lower-viscosity fluid (the displacing fluid) enters the porous medium at a constant rate, the ratio of gravitational and viscous forces increases and, therefore, gravity becomes progressively more important.

13.5

Models of Miscible Displacements in Hele-Shaw Cells

It is useful to consider miscible displacements in a Hele-Shaw cell of length L in which the two parallel plates are separated by a small distance b . For single-phase flow in the absence of dispersion and in the limits of small Reynolds number and small b/L , the governing equations are the continuity equation, $\nabla \cdot \mathbf{v} = 0$, and the Darcy's law. Because flow is confined between two parallel plates, one defines an equivalent permeability for the cell by $K_e = b^2/12$. However, these equations are the same as those of 2D incompressible fluids in a porous medium. As long as mixing due to dispersion is absent, the same analogy is also valid between viscous fingers in Hele-Shaw cells and 2D porous media.

With dispersion present, one must add the convective-diffusion (CD) equation to the set of the governing equations, and keep in mind that the viscosity μ_m of the mixed zone and the effective viscosities of the two fluids are now dependent upon the concentration C_s of the solvent (and, thus, that of the fluid to be displaced). Such concentration-dependent viscosities also give rise to nonlinear governing equations, the solution of which depends on the value of the Péclet number as well as on two other dimensionless groups that are

$$A = \frac{\mu_1 - \mu_2}{\mu_1 + \mu_2} = \frac{M - 1}{M + 1}, \quad (13.9)$$

$$B = \frac{g K_e (\rho_1 - \rho_2)}{V(\mu_1 + \mu_2)}, \quad (13.10)$$

where $M = \mu_1/\mu_2$. The most important aspects of a miscible displacement are its stability and efficiency. Thus, let us first outline the general approach to stability analysis of miscible displacements as we refer to it frequently in the subsequent sections. Later in this chapter, we present a more quantitative description of a stability analysis of miscible displacements, but for now, we restrict ourselves to a qualitative description.

The standard method of stability analysis of miscible (and immiscible) displacements may be summarized as follows. As the first step, the governing equations are introduced that represent the initial state of the system before it is perturbed and are called the *base-state* equations. Next, one introduces perturbations into the dependent variables of the model. The perturbations can be caused by many factors, including the heterogeneities of the pore space, or the viscosity contrast between the two fluids. Thus, one writes $\mathbf{v} = \bar{\mathbf{v}} + \mathbf{v}'$, $P = \bar{P} + P'$, and, $C = \bar{C} + C'$, where the bars indicate the unperturbed (base-state) or the mean values, and the primes represent the perturbations. The substitutions result in a set of equations that, when subtracted from the unperturbed equations, yields the governing equations for the perturbations.

Next, the governing equations for the perturbations are solved either analytically or numerically, and from the solution, the stability criteria are derived. Clearly, if the perturbations grow with time, then the displacement is unstable. At first, this may seem to be a straightforward exercise that can be carried out with essentially

no difficulty. However, the governing equations for the perturbations are nonlinear and difficult to solve, even numerically. Thus, the equations are linearized in order to make the computations more tractable. Doing so results in a *linear stability criterion* that is useful for the *onset* (short-time behavior) of instability, but cannot be used for predicting the long time behavior of the displacement process.

One common feature of linear stability theory is that one can decompose the initial perturbations into separate Fourier components so that the stability of each component can be investigated separately. This often simplifies the problem considerably. It also suggests that Fourier analysis (also known as spectral analysis) is a very convenient way to solve the perturbation equations. Fourier analysis also introduces the terminology of the wave theory and, thus, the stability criterion may be expressed in terms of perturbations that have wavelengths greater or less than a critical value.

Dispersion limits the wavelengths or frequencies of the Fourier components that can be unstable, which is why it usually has a positive effect on a miscible displacement. In the presence of dispersion, the problem is always time dependent, transport coefficients such as the viscosity and dispersion coefficients are concentration dependent, and one cannot determine a physical steady-state solution. Chouke was the first who analyzed the effect of dispersion, but his results appeared almost 30 years later in an appendix to the paper of Gardner and Ypma (1982). For now, we assume that the disturbances are of the form of normal modes proportional to $\exp(\omega t + i\alpha y)$. Chouke considered the case of a jump in viscosity, that is, a base-case solution in which the longitudinal dispersion is absent, and the concentration field can be divided into two distinct regions, one of which contains pure solvent, while the other contains pure displaced fluid. However, Chouke did allow both the longitudinal and transverse dispersion to act on the disturbances.

Chouke's result for ω , which essentially measures the rate of growth of the disturbances, is given by

$$\omega = \frac{1}{2} \left(A\alpha - \alpha^3 \text{Pe}^{-2} - \alpha \sqrt{\alpha^2 \text{Pe}^{-2} + 2A\alpha \text{Pe}^{-1}} \right), \quad (13.11)$$

where the cutoff wave number is given by

$$\alpha_c = \frac{1}{4} A \text{Pe}, \quad (13.12)$$

and ω is maximum when

$$\alpha_m = \frac{(2\sqrt{5} - 4)A\text{Pe}}{4}. \quad (13.13)$$

Thus, the Péclet number, $\text{Pe} = LV/D_m$, provides a physical mechanism for the introduction of a cutoff length scale. Equation (13.11) should be compared with the result in the absence of dispersion, that is, when $\text{Pe} \rightarrow \infty$, $\omega = (A + B)\alpha$, which is unphysical because it implies that smaller wavelengths are even more unstable than the larger ones. An equation such as Eq. (13.11) that relates ω to α is called a *dispersion relation*. The approach of introducing small perturbations was further

developed by Perrine (1961) and Wooding (1962). The latter treated the stability of a time-dependent base solution, and considered buoyancy-driven fingering. By expressing the disturbance quantities as an expansion in Hermite polynomials

$$H_n(x) = (-1)^n \exp(x^2) \frac{d^n}{dx^n} [\exp(-x^2)], \quad (13.14)$$

and truncating the expansion beyond the first term, Wooding obtained a dispersion relation that was qualitatively similar to Eq. (13.11). He also argued that all the disturbances will die out if dispersion is given enough time to act, but did not specify what constitutes “enough.”

Another approach was based on a macrostatistical method, first used by Scheidegger and Johnson (1961), in which the fingers were treated statistically. Thus, only the average cross-sectional areas occupied by the fingers were taken into account, and the shape and size of the individual fingers were neglected. Dougherty (1963), Koval (1963), and Perrine (1963) used such an approach and took into account the effect of dispersion. Koval’s model has been used widely in the petroleum industry, and is described in the next section.

Schowalter (1965) studied a fingering phenomenon in which both the density and viscosity variations were present. Although the governing equations do not allow a steady base solution, he assumed one anyway and obtained a dispersion relation and a cutoff wave number. Heller (1966) considered horizontal miscible displacement in a rectangular system and studied the early growth or decay of the perturbations using a Fourier analysis. He also included the effect of dispersion and approximated the profiles by straight-line segments (which he called *ramp-shaped* profiles), and obtained the dependence of the growth exponent on the wave number. However, the assumption of straight-line segments for the profiles limited the applicability of his results.

Peters *et al.* (1984) considered a miscible displacement process, took into account the effect of dispersion, and performed a Fourier analysis to derive a linear stability criterion. Assuming the perturbations to be proportional to $\exp(\omega t + \alpha_x x + \alpha_y y)$, they proposed that the displacement is stable if

$$\frac{1}{8\pi^2} (\alpha_x^2 + \alpha_y^2) < \frac{1}{4\pi} \sqrt{\frac{K_e \frac{g d\rho}{dC} - \frac{V d\mu}{dC}}{\phi \delta_t \bar{\mu} D_T}} \quad (13.15)$$

where δ_t is the length of the mixing zone. The right side of Eq. (13.15) defines a critical frequency or wave number ω_c , which should be compared with Eq. (13.11). Tan and Homsy (1986) studied the same problem for the case of no density difference, and one in which the concentration dependence of the viscosity in the mixed zone was given by $\mu_m(C) = \exp(-C \ln M)$. The flow was rectilinear and the domain was unbounded. Both isotropic ($D_L = D_T$) and anisotropic dispersion were considered, and the quasi-steady state assumption was made. They showed that for the isotropic case, Chouke’s result is correct in the sense that it correctly predicted the magnitude of the growth rates and the preferred wave numbers. They also found, however, that at longer times, dispersion causes a shift to larger wave

lengths and stabilizes the flow to some extent. Tan and Homsy (1986) also showed that the transverse dispersion causes a shift to smaller length scales, as expected.

Hickernell and Yortsos (1986) considered the case in which the amount of the solvent injected into the system varied with time, resulting in a spatially-varying mobility profile. They ignored the effect of the dispersion and showed that a finite thickness of the zone of viscosity variations provides a cutoff scale. Chang and Slattery (1986) showed that when there is a steep change in the concentration and $M > 1$, the displacement can be unstable at the injection boundary. However, if the concentration is changed sufficiently slowly with the time at the entrance to the system, the displacement will be stable to small perturbations, *regardless of the value of M* .

In some situations, mixing by dispersion plays a role similar to that of surface tension in immiscible displacements. As we describe in Chapter 14, surface tension is responsible for *tip splitting* in the fingers, which is the main mechanism of pattern formation in many phenomena, such as crystal growth. Mixing by dispersion can cause a similar phenomenon in miscible displacements. In tip splitting, the tip of a finger becomes unstable and splits into two branches that compete with each other for further growth. An example is shown in Figure 13.2. The experiments of Wooding (1969) in a Hele-Shaw cell in the presence of buoyancy forces are indicative of this phenomenon. In his experiments, the transverse dispersion causes lateral *spreading* of the fingers, as expected. Because of the spreading, however, the tips of the fingers may become unstable since their typical breadth exceeds the cutoff length scale, which is also set by the transverse dispersion. Due to this instability, tip splitting can occur if Pe exceeds a critical value that, according to Tan and Homsy (1987), depends on both A and M . For example, at $M = e^3$, $250 < (Pe/R_0)_{\text{critical}} < 300$ if dispersion is isotropic (i.e., $D_L = D_T$), where R_0 is the aspect ratio of the cell. Note that if the transverse dispersion is weak, then tip splitting may not happen at all because before it can start, one must have spreading of the fingers, which can happen only if the transverse dispersion is strong enough.

Numerical simulations of miscible displacements in rectilinear flows with weak dispersion (high values of Pe) are particularly difficult. The main reason for the difficulty is that for large values of Pe , viscous fingering can occur on many scales, and there is no unique power of Pe with which one can rescale all the lengths. The first attempt in this direction was made by Peaceman and Rachford (1962) using a finite-difference (FD) method. The attempt was not, however, successful due to the large numerical errors that dominated the solution. Since their pioneering attempt, considerable efforts have been dedicated to the problem. Zimmerman and Homsy

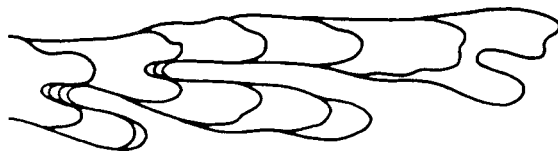


Figure 13.2 Tip splitting in viscous fingering in a Hele-Shaw cell (after Liang, 1986).

(1991), for example, proposed a method that appears to tackle the problem to a large extent. They used a 2D discrete Hartley transformation that, for an arbitrary function $g(x, y)$, is defined by

$$G(\alpha_x, \alpha_y) = \frac{1}{\sqrt{N_x N_y}} \sum_x \sum_y \text{cas} \left(\frac{2\pi x \alpha_x}{N_x} + \frac{2\pi y \alpha_y}{N_y} \right) g(x, y), \quad (13.16)$$

where N_x and N_y are the number of collocation points (the points at which the discrete Hartley transformation of the function is computed), α_x and α_y are the wave numbers in the longitudinal and transverse directions, respectively, and $\text{cas}(x)$ is the so-called “cosine and sine” function formed by summing the cosine and sine of its argument. The advantage of using the Hartley transformation is that it can easily be inverted because it is its own inverse. An efficient method for computing a 2D discrete transformation via $N_x + N_y$ discrete fast Hartley transformations was devised by Bracewell *et al.* (1986). The Hartley transformation recasts the system of governing partial differential equations into an ordinary differential equation for dC/dt . The resulting equation is then integrated and transformed back into the (x, y) space. Zimmerman and Homsy (1991) used the Taylor–Aris result (see Chapter 11) to relate D_L to Pe for dispersion in flow between two parallel plates (Hele-Shaw cell), and assumed that $D_T = D_m$. As such, the results are valid only for Hele-Shaw-like systems, although the authors claimed that they are also valid for porous media.

Over a wide range of the Péclet number Pe , Zimmerman and Homsy (1991) observed a variety of phenomena, including spreading, tip splitting, shielding, and pairing. In a shielding process, one finger gets ahead of its neighbors and grows explosively. Eventually, the tip of the finger spreads out and *shields* the neighboring fingers. An example is shown in Figure 13.3. Pairing is a phenomenon by which pairs of fingers join and form a larger finger. This phenomenon has been observed with both isotropic ($D_L = D_T$) and anisotropic dispersion, although as far as porous media flows are concerned, isotropic dispersion is not relevant. Pairing may also happen in immiscible fingering, as was shown by Tryggvason and Aref (1985) and Kessler and Levine (1986). According to Tan and Homsy (1987), pairing occurs due to unequal cross-flow about neighboring fingers, which allows a finger to spread and shield the growth of the neighboring finger, and results in its eventual

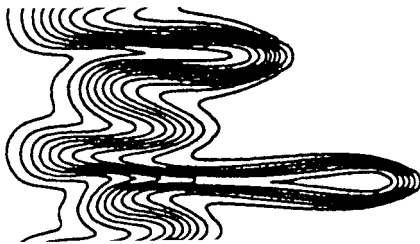


Figure 13.3 Shielding phenomenon in which one finger spreads out and shields all other fingers (after Zimmerman and Homsy, 1991).

collapse. If tip splitting is absent, then pairing eventually results in the reduction of the number of fingers to one or two. The study of Zimmerman and Homsy (1991) also indicated great sensitivity of the complex 2D fingering patterns that evolve to the size of the initial noise. However, when an averaging was performed over the cross-sectional area, it was found that the 1D average concentration profiles were similar for large Pe and D_L/D_m . This implies that it may be possible to describe nonlinear viscous fingers by a 1D model that is invariant with respect to Pe and D_m/D_L , for large Pe and small D_m/D_L . In a sense, this is the same as the aforementioned idea of Scheidegger and Johnson (1961). It is also similar to the work of Fayers (1988) who constructed an approximate 1D model with adjustable parameters that had clear physical meaning. Koval (1963) and Todd and Longstaff (1972) also constructed empirical 1D models that could accurately predict the evolution of the average concentration profile. Their models are described shortly. Finally, Christie and Bond (1987) developed a numerical method for the evolution of both linear and nonlinear fingers using a FD method for the longitudinal direction and Fourier decomposition in the transverse direction.

The effect of the cell geometry is significant. Wilson (1975) and Paterson (1985) studied miscible viscous fingering in a radial Hele-Shaw cell, both theoretically and experimentally. Paterson (1985) ignored dispersion and obtained the following relation for ω

$$\omega = Am - 1, \quad (13.17)$$

where m is a discrete azimuthal wave number. Equation (13.17) indicates that there is no cutoff azimuthal wave number. Thus, Paterson (1985) suggested that in a radial Hele-Shaw cell and in the absence of the dispersion, fingers can form on *all* length scales down to the size of the gap between the plates. Based on a heuristic argument, Paterson (1985) estimated that the cutoff wavelength is approximately $4b$. Figure 13.4 shows the pattern of miscible viscous fingers in a radial Hele-Shaw cell. While his experimental results appear to agree with his rough estimate of the cutoff wavelength, Paterson's argument is not expected to hold for the general case in which the dispersion is present because, as discussed earlier, the transverse dispersion (however small in a Hele-Shaw cell) helps the fingers to spread, which leads to tip splitting. Thus, fingers cannot form down to the scale b .

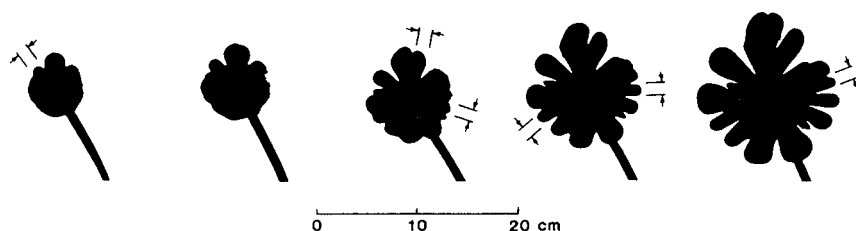


Figure 13.4 Viscous fingers in a radial Hele-Shaw cell. The gap between the plates is $b = 0.3$ cm, and the exposures are, from left to right, at times $t = 12, 17, 21$, and 31 s. The indicated distance is $4b$ (after Paterson, 1985).

A different formulation of finger formation in Hele-Shaw cells was presented by Bogoyavlenskiy (2001) based on a generalization of the diffusion-limited aggregation model that will be described shortly. For the latest developments on viscous fingers in Hele-Shaw cells, see Logvinov *et al.* (2010).

13.6

Averaged Continuum Models of Miscible Displacements

As mentioned earlier, since the 1950s, there have been many experimental studies of miscible displacements in porous media, most of which were unconsolidated. In particular, Slobod and Thomas (1963) and Perkins *et al.* (1965) used X-ray techniques to make visualization studies of viscous fingers. Normally, one can observe viscous fingers over length scales that are several times larger than the typical pore size. Since in such experiments the transverse Pe (i.e., the Péclet number that is based on D_T , instead of D_m) is relatively low, finger growth is probably due to the spreading phenomenon. A careful examination of the pictures taken during the experiments of Slobod and Thomas (1963) and Perkins *et al.* (1965) shows no tip splitting. Habermann (1960)'s experiments, however (that are similar to those shown in Figure 13.1), and those of Mahaffey *et al.* (1966) do indicate tip splitting due to shielding, similar to those in a Hele-Shaw cell already described.

The dominant length scale in all of such experiments is much larger than a typical pore size. However, if one carries out experiments in a porous medium in which the Pe increases with decreasing length scales, one can no longer claim that the dominant length scale is much larger than the typical pore size. An example is the work of Paterson *et al.* (1982) who studied miscible viscous fingers in packed beds, and obtained viscous fingers with a very open structure and many thin branches (see also below). This indicates that, for a given pore space, there is a crossover between the continuum and discrete description of miscible displacements. An important and interesting question is the shape of the viscous fingers for the case in which there is no mechanism for the creation of a cutoff length scale, for example, in the absence of dispersion.

Let us also point out that Juanes and Blunt (2006) presented a number of analytical solutions for a model of viscous fingering for three-component, two-phase, first-contact miscible flows, and in particular, for the water-alternating-gas floods. However, obtaining analytical solutions for the more general problems involving miscible displacements is very difficult. Thus, one must search for alternative ways of analyzing the problem.

One approach to describing miscible displacements and fingering phenomena in a porous medium is based on averaged continuum equations. The method can describe any instability that is smooth on the length scale over which the continuum description is applicable. In the strict absence of dispersion (i.e., the limit $Pe \rightarrow \infty$, with Pe being based on D_L or D_T and not D_m), fingers form at all length scales with growth rates that increase with decreasing length scale. This means that a length scale is reached at which a continuum description is no longer appro-

priate, and one must develop a pore-scale model. Such models will be described below. In this case, the initial-value problem that describes the phenomenon is ill-posed, but one can seek solutions that contain discontinuities. Since dispersion is absent in this case, there will be a step jump in the viscosity profiles (from that of the displacing fluid to the displaced one). As a result, the pressure satisfies the Laplace equation, and the pressure and fluid fluxes are continuous across the front separating the displaced and displacing fluids. One may have all types of singularities in the solution, with different nonuniformities appearing in various boundary-value problems. Shraiman and Bensimon (1984) and Howison (1986) studied such phenomena, but they are beyond the scope of this book. Two popular 1D semi-empirical continuum models of miscible displacements are those due to Koval (1963) and Todd and Longstaff (1972).

13.6.1

The Koval Model

Koval recognized that the central feature of the physics of viscous fingers is linear growth of the fingers' length with time. Thus, to ensure that this is a feature of his model, Koval cast the problem as a hyperbolic transport equation similar to the more familiar Buckley–Leverett equation of two-phase flow to be studied in Chapter 14. In Koval's model the displacing fluid is assumed to travel at a constant, characteristic velocity v . If the flow is linear, then

$$v = \left(\frac{dx}{dt} \right)_s = \frac{Q}{\mathcal{A}\phi} \left(\frac{df_s}{dS} \right), \quad (13.18)$$

where S is the saturation of the displacing fluid (i.e., its volume fraction in the pore space), Q is the volumetric flow rate, $\mathcal{A}$ is the cross-sectional area, and f_s is the volume fraction of the solvent (the displacing fluid) in the flowing fluid. Koval made an analogy between miscible and immiscible displacements. For an immiscible displacement of oil by water, the Buckley–Leverett equation, when gravity and the capillary pressure are negligible, relates f_w , the fractional flow of water, to the permeabilities K_w and K_o of the water and oil phases. Thus,

$$f_w = \frac{1}{1 + \left(\frac{K_o}{K_w} \right) \left(\frac{\mu_w}{\mu_o} \right)}. \quad (13.19)$$

Koval argued that permeability to either oil or the displacing fluid can be expressed as the total permeability K_e multiplied by the average saturation of each fluid. Thus, if viscous fingering is the dominant phenomenon, one can write

$$f_s = \frac{1}{1 + \left(\frac{1}{H_K} \right) \left(\frac{\mu_{es}}{\mu_{eo}} \right) \left(\frac{1-S}{S} \right)}, \quad (13.20)$$

where μ_{es} and μ_{eo} are the *effective* viscosities of the solvent and oil (the displaced fluid), respectively, and H_K is called the Koval heterogeneity index, introduced in

Section 5.2.5, that characterizes the inhomogeneity of a porous medium. To correlate H_K with some measurable quantity, Koval defined a homogeneous porous medium as one in which the oil recovery, after one pore volume of the solvent has been injected into the medium is 99%. Thus, for a homogeneous porous medium, $H_K = 1$, whereas for any other porous medium for which the recovery is less than 99% $H_K > 1$. Empirically, H_K and the recovery appear to be linearly related. Based on experimental data, Koval (1963) also suggested the following expression for the effective viscosity ratio,

$$M_e = \frac{\mu_{eo}}{\mu_{es}} = \left[0.78 + 0.22 \left(\frac{\mu_o}{\mu_s} \right)^{\frac{1}{4}} \right]^4, \quad (13.21)$$

which is similar to Eq. (13.1). Figure 13.5 presents the correlation between H_K and the recovery.

The solution to Eqs. (13.18) and (13.20) at solvent breakthrough (i.e., at the point where the solvent forms a sample-spanning cluster) is

$$V_{BT} = \frac{1}{M_e H_K}, \quad (13.22)$$

where V_{BT} is the injected pore volume of the solvent at the breakthrough. The fractional pore volume of the recovered oil f_o is predicted to be

$$f_o = \frac{2\sqrt{M_e H_K V_{BT}} - V_{BT} - 1}{M_e H_K - 1}, \quad (13.23)$$

and the length L_f of the region in which fingering has occurred is given by

$$L_f = \left(M_e H_K - \frac{1}{M_e H_K} \right)_{x_m}, \quad (13.24)$$

where x_m is the mean displacement distance.

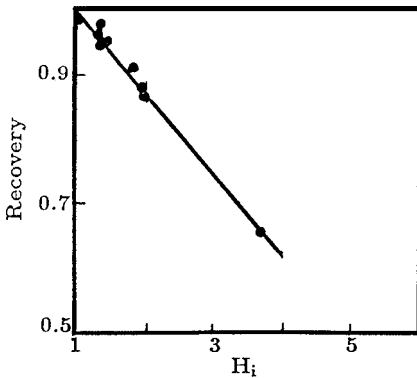


Figure 13.5 Dependence of the heterogeneity index H_K on the recovery based on matched viscosity experiments (after Koval, 1963).

Figure 13.6 compares the predictions of Koval's model with the experimental data of Blackwell *et al.* (1959). The agreement is very good. It can be shown that Koval's model can also be used for predicting miscible displacements and their recovery efficiency in Hele-Shaw cells. However, despite its apparent success, Koval's model suffers from two fundamental shortcomings.

1. Due to the empirical nature of Eq. (13.21), it is not clear why it should be an adequate representation of the viscosity ratio, which in effect measures the effect of mixing during a miscible displacement.
2. The heterogeneity index H_K is inadequate for characterizing the structure of a porous medium. The correlation shown in Figure 13.5 is based on laboratory data, whereas field applications of miscible displacements involve large-scale porous media that are characterized by long-range correlations and very broad spatial variations of the permeability (see Chapter 5), the effect of which cannot possibly be included in the correlation shown in Figure 13.5.

13.6.2

The Todd–Longstaff Model

In the model developed by Todd and Longstaff (1972), the average concentration $\bar{C}_s$ of the solvent is described by

$$\frac{\partial \bar{C}_s}{\partial t_D} + \frac{\partial \bar{f}_s}{\partial x_D} = 0, \quad (13.25)$$

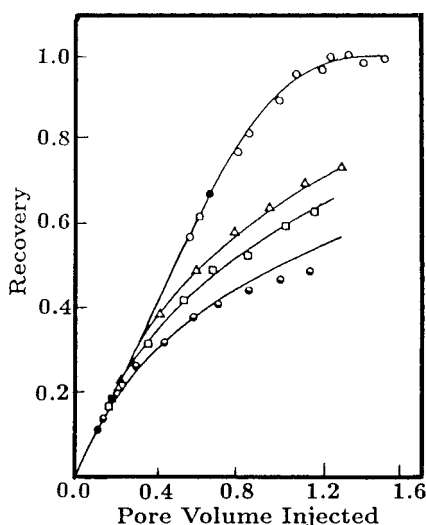


Figure 13.6 Comparison of the predictions of Koval's model (curves) with the experimental data of Blackwell *et al.* (1959). The results are, from top to bottom, for $M = 5, 86, 150$, and 375 (after Koval, 1963).

where $\bar{f}_s$ represents the average of f_s and is a function of $\bar{C}_s$, and x_D and t_D are dimensionless distance and time, respectively. Equation (13.25) represents the limit of the CD equation in which the dispersion term has been neglected. Todd and Longstaff assumed that

$$\mu_{eo} = \mu_o^{1-\zeta} \mu_m^\zeta, \quad (13.26)$$

with a similar expression for μ_{es} , where μ_m is given by Eq. (13.1) in which $\bar{C}_s$, instead of C_s , is used, and $\zeta \simeq 2/3$. $\bar{f}_s$ is given by

$$\bar{f}_s = \frac{\bar{C}_s}{\bar{C}_s + M_e^{-1}(1 - \bar{C}_s)}. \quad (13.27)$$

The solution of Eqs. (13.25) and (13.27) indicate that the average concentration $\bar{C}_s$ moves with a speed $d\bar{f}_s/d\bar{C}_s$. Therefore, the leading edge of the finger, where $\bar{C}_s = 0$, moves at a speed $d\bar{f}_s/d\bar{C}_s = \mu_{eo}/\mu_{es}$, whereas the trailing edge, where $\bar{C}_s = 1$, moves at a speed $d\bar{f}_s/d\bar{C}_s = \mu_{es}/\mu_{eo}$.

To investigate the effect of the heterogeneities on the performance of the model, Todd and Longstaff fitted the predictions of their model to the experimental data for three systems, namely, a Hele-Shaw cell, a bead pack, and a consolidated sand pack, using ζ as the adjustable parameter. The Hele-Shaw cell does not contain any heterogeneity, whereas the two porous media contain very different heterogeneities. Figure 13.7 compares the fitted predictions with the data, and it should be clear that it is impossible to predict all the three sets of data with a single value of ζ . Note also the difference between the Hele-Shaw results and those of the

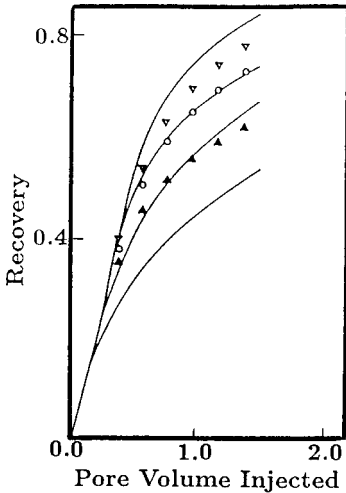


Figure 13.7 Comparison of the predictions of Todd–Longstaff model (curves) with the experimental data for a Hele-Shaw cell (solid triangles), bead pack (circles), and consoli-

dated sandpack (open triangles). The results are, from top to bottom, for $\zeta = 1, 2/3, 1/3$, and 0, and $M = 10$ (after Todd and Longstaff, 1972).

consolidated sandpack. It has been suggested that a good fit of the model's predictions to the experimental data of Blackwell *et al.* (1959) is obtained if one sets $\zeta = 1 - \log M_e / \log M$, where $M = \mu_o / \mu_s$. We may conclude that the model of Todd and Longstaff suffers from the same shortcomings as those of Koval's model. Due to their simplicity, however, both the Koval and Todd–Longstaff models have been used heavily in the petroleum industry.

More recent models that are more or less based on the same type of averaged continuum equations are those of Vossoughi *et al.* (1984), Newley (1987), Fayers (1988), Odeh and Cohen (1989), and Fayers *et al.* (1990). Although such models may provide an adequate fit to the production/effluent data for either 1D or 2D porous media, they often lead to quite different predictions of the pressure field during the unstable displacement process. In the evaluation of the averaged models of viscous fingering, their performance in the 2D model, where viscous instability occurs, has been considered (Newley, 1987; Fayers *et al.*, 1990). To distinguish between such models experimentally, it is necessary to measure the pressure field directly during unstable miscible displacements, an extremely difficult task to carry out for either 2D linear or other geometries (see, however, Sorbie *et al.*, 1995). Alternatively, the computed pressure distribution may be used as “experimental” data for assessing the predictions of the averaged equations. Such calculations should, however, be validated by comparison with experimental results.

13.7

Numerical Simulation

Numerical simulations of unstable miscible displacements have been carried out for a long time. The main difficulty in such simulations lies in the correct reproduction of the wide range of the relevant length and time scales that typically characterize such phenomena. At the smallest length scale, the size of the smallest grid block is determined by the action of physical diffusion or dispersion, whereas the reservoir linear size, or the distance between the wells, determines the large scales. As already pointed out, Peaceman and Rachford (1962) developed a FD algorithm for computing an unstable rectilinear miscible displacement. Due to the low order of their numerical algorithm, a fingering instability did not develop on its own, but had to be triggered artificially by imposing small random spatial variations in the permeability. Over the nearly five decades following their work, a host of novel numerical approaches for the simulation of miscible displacements has been introduced and tested on problems of varying degrees of difficulty. In what follows, we briefly describe some of the methods.

13.7.1

Finite-Element Methods

For a comprehensive discussion of the efforts up until 1983, see the review by Russell and Wheeler (1983). Douglas *et al.* (1984) presented the results of computer

simulations of unstable miscible displacements for one quarter of the five-spot geometry. They used a self-adaptive finite-element (FE) method; see also Bell *et al.* (1985). Darlow *et al.* (1984) described the so-called mixed FE methods, which offer certain computational advantages particularly for strongly heterogeneous porous media by solving for pressure and velocity *simultaneously*. Ewing *et al.* (1984) analyzed a modified method of characteristics for handling the governing equation for the concentration.

Ewing *et al.* (1989) performed more detailed FE simulations of miscible displacements in anisotropic and heterogeneous porous media in one quarter of five-spot geometry using computational grids of up to size 100×100 . They reported that, typically, viscous fingering instability was much stronger than the permeability-related effects. In general, the relative importance of the two effects is expected to depend on the viscosity ratio and the degree of heterogeneity of the porous medium. Comprehensive reviews of the the FE methods as well as related work are provided by Ewing and Wang (1994), along with a discussion of the methods in light of recent developments in computer architectures; see also Arbogast *et al.* (1996).

13.7.2

Finite-Difference Methods

Conventional FD discretizations have also been used for obtaining improved physical understanding of miscible displacements; see, for example, Giordano and Salter (1984), Giordano *et al.* (1985), Christie and Bond (1987), Christie (1989), Bradtvedt *et al.* (1992), Fayers *et al.* (1992), and Christie *et al.* (1993) as well as Sorbie *et al.* (1995) and Zhang *et al.* (1996). The overall accuracy of the FD methods are typically of second order, but often they still suffer from significant numerical dispersion. In terms of formal accuracy, an important seminal work was that of Leventhal (1980), who applied the fourth-order operator compact implicit family of the FD schemes (see, for example, Shukla and Zhong (2005)) to 1D, two-phase immiscible water-flood problems, and demonstrated a significant reduction of the adverse effect of numerical diffusion. Tchelepi and Orr (1993, 1994) minimized the amount of numerical diffusion by employing a particle tracking technique (Araktingi and Orr, 1988; see below), although the solution of the accompanying pressure equation still resulted in some artificial smoothing.

13.7.3

Streamline Method

In this approach, the flow problem is decoupled into a set of 1D problems solved along streamlines which reduces the simulation time and suppress the numerical dispersion. The obvious advantages of the method have increased its wide application and fast commercialization. However, compared with the conventional FD simulations, the streamline-based simulations usually only account for relatively simple physics. Such simulators are limited to production scenarios where the effect of gravity can be neglected. Given that compositional simulations are much

more time consuming, it is of great interest to speed up the compositional simulators using a streamline method.

Thiele *et al.* (1995) proposed a compositional simulation approach to investigate displacements in 2D heterogeneous reservoirs. Thiele *et al.* (1997) combined a 3D streamline method with a 1D two-phase numerical solver and developed a streamline-based 3D compositional reservoir simulator. Jessen and Orr (2002) developed a 3D compositional reservoir simulator that maps the analytical solution of a 1D two-phase multicomponent gas injection along each streamline, resulting in the simulator being orders of magnitude faster and completely free of numerical dispersion. A limitation of the analytical mapping method is, however, that it is only applicable to the uniform initial condition.

More recently, Yan *et al.* (2004) developed a three-phase compositional streamline simulator by integrating a 1D numerical solver. The numerical solver is optimized for calculating the efficiency, and various descriptions of phase equilibrium between hydrocarbon and water as well as the gravity effect are included by the operator-splitting (OS) method, which relies (Bradtvedt *et al.*, 1996; Batycky *et al.*, 1996; Batycky, 1997) on the consistency of treating the convective flux independently of gravity flux within a given time step of the simulation. For small time steps, the OS approximation is fairly accurate, whereas large time steps may lead to significant errors. Jessen and Orr (2004) used the OS technique to develop a compositional simulator suitable for displacement processes with significant gravity segregation.

13.7.4

Spectral Methods

To overcome the limitation in the accuracy of many of the above grid-based methods, significantly more accurate spectral methods have also been developed (see, for example, Tan and Homsy, 1988; Zimmerman and Homsy, 1992a,b; Rogerson and Meiburg, 1993a,b). Such methods possess superior accuracy for smooth flows, that is, for flows without discontinuities and singularities (Gottlieb and Orszag, 1977), and avoid problems associated with numerical diffusion. In addition, the spectral simulations are carried out for the governing equations that are formulated in terms of the vorticity and the stream functions. Since one no longer needs to solve for the pressure distribution, such formulation of the governing equations often leads to higher computational efficiency. Moreover, such a formulation offers the advantage that it satisfies the conservation of mass identically. Thus, such methods allow for detailed investigation of mobility- and gravity-driven fingering processes in rectilinear displacements at relatively low levels of physical dispersion.

13.8

Stability Analysis

Theoretically, miscible displacements are very efficient EOR processes. Because capillary forces, which are usually responsible for oil entrapment, are absent, the displacement can potentially be 100% efficient. In practice, however, complete recovery is usually not realized due to the instability phenomena that leads to macroscopic fingering of the solvent. Over the past several decades, many stability analyses of miscible displacements have been carried out (Perrine, 1963; Dumore, 1964; Schowalter, 1965; Heller, 1966; Gardner and Ypma, 1982; Lee *et al.*, 1984; Peters *et al.*, 1984). Among the unsolved problems is the question of what causes the disturbances that propagate as the fingers. It is widely believed that the heterogeneities of porous media are the source of the disturbances. A pore space is not, however, necessary for the development of the fingers. Indeed, as described earlier, pioneering studies of viscous fingering were carried out using the Hele-Shaw cell, the uniformity of which should preclude any cell-related source of instability. This is actually seen in the early time radial behavior at the inlet of a radial Hele-Shaw Cell (Kopf-Sill and Homsy, 1988; Meiburg and Homsy, 1988). At the same time, explanation of fingering in Hele-Shaw cells is typically based on the assumption of the presence of a perturbation in the system. Moreover, some studies (Christie, 1989; Fanchi and Christianson, 1989; Fanchi, 1990) suggest that nonlinear dynamics of a miscible displacement may provide an alternative source for the disturbances in porous media.

Kelkar and Gupta (1988) reported that they were unable to initiate a finger without introducing some type of perturbation, for example, a permeability variation or a concentration perturbation. Araktingi and Orr (1988) introduced randomness in their model (see below). Such studies illustrate the approach taken by many researchers in the field: Identify a parameter in the system with values that exhibit some degree of spatial randomness. The randomness then becomes the source of the perturbation. A problem with this approach is its dependence on the *ad hoc* incorporation of a probabilistically-distributed variable.

As already described in Chapter 11, dispersion coefficients depend on the mean flow velocities, with the precise form of their dependence depending on the value of the Péclet number Pe . For simplicity, however, we assume that D_L and D_T are related to the flow velocities V_x and V_y through the following relations (Bear, 1972):

$$D_L = D_m + \alpha_L |V| + \frac{(\alpha_L - \alpha_T) V_x^2}{|V|}, \quad (13.28)$$

$$D_T = D_m + \alpha_T |V| + \frac{(\alpha_L - \alpha_T) V_y^2}{|V|}, \quad (13.29)$$

which describe the longitudinal and transverse dispersion coefficients in terms of the corresponding dispersivities α_L and α_T as well as the molecular diffusivity D_m . For convenience, we shall follow the notation in Yortsos and Zeybek (1988). The governing equations are the CD and the continuity equations for an incompressible

fluid, coupled with Darcy's law:

$$\phi \frac{\partial C}{\partial t} + V_x \frac{\partial C}{\partial x} + V_y \frac{\partial C}{\partial y} = \frac{\partial}{\partial x} \left(D_L \frac{\partial C}{\partial x} \right) + \frac{\partial}{\partial y} \left(D_T \frac{\partial C}{\partial y} \right), \quad (13.30)$$

$$\frac{\partial V_x}{\partial x} + \frac{\partial V_y}{\partial y} = 0. \quad (13.31)$$

Implicit in the above continuum description is the assumption that the local Péclet number VL/D_m is small.

The base-state solution that corresponds to a constant injection rate in a rectilinear flow geometry is given by the well-known diffusive profile (see Chapter 11)

$$\bar{C} = \frac{1}{2} \operatorname{erfc} \left[\frac{\xi}{2\sqrt{t}} \right], \quad (13.32)$$

$$\frac{\partial \bar{P}}{\partial \xi} = -\frac{1}{\lambda(\bar{C})} \quad (13.33)$$

where $\xi = (x - Vt)/L$ is a moving coordinate, λ denotes a normalized mobility (inverse of viscosity), and all the lengths are scaled with D_{L0}/V , with D_{L0} being the base-state dispersion coefficient. Writing $C = \bar{C} + C'$, $V = \bar{V} + V'$, and $P = \bar{P} + P'$, and using normal modes for the concentration and flow rate, respectively

$$(C', V'_x) = (\Sigma, \Phi) \exp(\omega t + i\alpha y), \quad (13.34)$$

the following equations are obtained.

$$\Sigma_{\xi\xi} - (\epsilon\alpha^2 + \omega)\Sigma = \Phi C_{\xi} - L_c(\Phi C_{\xi})_{\xi}, \quad (13.35)$$

$$\lambda(\lambda^{-1}\Phi_{\xi})_{\xi} - \alpha^2\Phi = -\alpha^2 R\Sigma, \quad (13.36)$$

where the concentration dependence of the mobility is taken to be (Tan and Homsy, 1986)

$$\lambda(C) = \exp(RC), \quad (13.37)$$

and $R = \ln M$. The subscripts denote derivatives with respect to the variables. Two important terms, ϵ and L_c

$$\epsilon = \frac{D_m + \alpha_T V}{D_m + \alpha_L V}, \quad (13.38)$$

$$L_c = \frac{\alpha_L V}{D_m + \alpha_L V}, \quad (13.39)$$

appear in Eq. (13.35). ϵ is a measure of flow-induced anisotropic dispersion and is a characteristic of porous media, while L_c is a measure of the contribution of mechanical dispersion to the total dispersion. It must be stressed that in the works of Chouke and Tan and Homsy (1986), the term containing L_c in Eq. (13.35) is

absent, thereby restricting their conclusions to essentially constant (although still anisotropic) dispersion. Zimmerman and Homsy (1991) used velocity-dependent dispersion coefficients, although, as mentioned earlier, their functional forms were different from Eqs. (13.28) and (13.29).

Based on the discussion presented in the previous sections, one expects the onset of instability and related features to be dictated by the sharpest mobility contrast, namely, those associated with a step concentration profile, which also allow for an analytical solution given by (Yortsos and Zeybek, 1988)

$$\alpha R \left[1 + L_c \gamma_0 \tanh \left(\frac{R}{2} \right) \right] = 2\gamma_0(\alpha + \gamma_0) , \quad (13.40)$$

where

$$\gamma_0 = \sqrt{\epsilon \alpha^2 + \omega} > 0 . \quad (13.41)$$

In general, the solution of Eq. (13.35) leads to parabolic-like profiles, examples of which are shown in Figure 13.8. The case $L_c = 0$ corresponds to the result of Tan and Homsy (1986): for an unfavorable mobility ratio ($R > 0$), large wavelengths are unstable, while a strong stabilization due to the transverse dispersion is exerted at smaller wavelengths. A cutoff wave number can be identified

$$\alpha_c = \frac{R}{2(\epsilon + \sqrt{\epsilon})} . \quad (13.42)$$

As expected, α_c increases with increasingly unfavorable mobility, and with an increase in the ratio of the longitudinal and transverse dispersion coefficients, $1/\epsilon$. However, the limits of the continuum description should be kept in mind. The size of the most unstable disturbance scales with the characteristic length of the porous medium that, for large enough flow rates, becomes equal to the dispersivity α_L , which is normally a multiple of the typical pore size (or the length scale of the heterogeneities). It is apparent that a possible conflict may develop between the above result and the continuum description, precluding meaningful predictions over scales of the order of the microscale.

While the case $L_c = 0$ leads to the expected results, a distinct sensitivity develops as L_c takes nonzero values (see Figure 13.8). This effect is present only due to the velocity dependence of the dispersion coefficients, and is best quantified in terms of the following combination

$$B_c = \frac{1}{2} R L_c \tanh \left(\frac{R}{2} \right) - \sqrt{\epsilon} - 1 . \quad (13.43)$$

The following results may then be shown (Yortsos and Zeybek, 1988):

1. When $B_c < 0$ (which is *always* the case if $L_c = 0$), at small enough viscosity ratio and for $L_c \neq 0$, the cutoff wave number is finite

$$\alpha_c = \frac{R}{2\sqrt{\epsilon}} (-B_c) , \quad (13.44)$$

although it increases as L_c or B_c does.

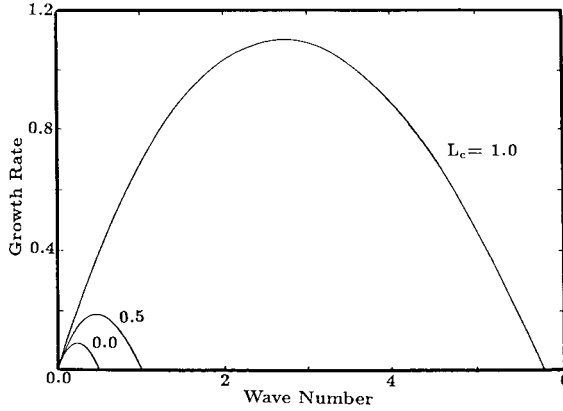


Figure 13.8 Step-profile results for the growth rate (after Yortsos and Zeybek, 1988).

2. On the other hand, when $B_c > 0$, a finite cutoff does not exist, with the rate of growth increasing indefinitely at large α as

$$\omega \sim B_c(B_c + 2\sqrt{\epsilon})\alpha^2 > 0. \quad (13.45)$$

Clearly, such is the case for a sufficiently high (but finite) mobility contrast as long as $L_c \neq 0$, as shown in Figure 13.8. This unexpected and rather remarkable result was obtained on the basis of a step base state, which is subject to a singular behavior in the large (as well as in the small) α region.

To better understand the results, Yortsos and Zeybek (1988) attempted a more rigorous asymptotic analysis valid for the base states near a step profile, namely, for

$$\bar{C} = \frac{1}{2} \operatorname{erfc}(c\xi), \quad (13.46)$$

where $c \gg 1$. Their analysis showed that the step profile prediction, inequality (13.46), is invalid at large α when $B_c > 0$ and, in fact, a cutoff wave number does exist. However, the latter was found to increase monotonically and without bound as c increases, namely, as the profile becomes step-like, provided that $B_c > 0$. Thus, the essential prediction that qualitatively different instability behavior is obtained by changing the sign of B_c remains intact. Most of the above results were confirmed by the experimental study of Bacri *et al.* (1991).

Thus, due to the dependence of the dispersion coefficients on the flow velocity, and for mobility ratios that exceed a critical value dictated by the given process conditions, a miscible displacement is predicted to be unstable *at all the wavelengths*. Under such conditions, there is no finite preferred mode and, in fact, the continuum description is ill-posed and breaks down. This remarkable result raises serious doubts about our ability to describe the conditions for the onset of instability in miscible displacements. Recall that this result is the outcome of several hypotheses, including the validity of a continuum description, with the dispersion formulated by a CD equation and the dispersion coefficients represented by Eqs. (13.28)

and (13.29). If these predictions are to be taken seriously, the breakdown of the continuum hypothesis beyond a finite M calls for an alternative description (to the present CD equation-based description), and vice versa.

One concludes that at least for large enough M , the present description of miscible displacements is inadequate, particularly at the early and the most important stage of the process. Thus, although Zimmerman and Homsy (1991) and others used a CD equation, one needs to establish that their formulation is actually applicable to the early stages of the growth of the fingers. The stability analysis clearly demonstrates the significance of anisotropic dispersion, that is, the inequality of D_L and D_T . We should also keep in mind that the results so far are limited to linear fingers, and nonlinear fingers require full numerical simulations of the governing equations, provided that the proper forms of velocity-dependence of the dispersion coefficients are used.

The above linear stability analysis is valid for short times. Hence, it should be extended to longer times into the nonlinear regime in order to not only check the accuracy of the linear stability analysis, but also understand the difference(s) between the linear and nonlinear regimes. There are also other factors, not considered in the above analysis, whose effect may be important to miscible displacements. Hence, Riaz and Meiburg (2003) carried out linear stability analysis for axial and helical perturbation waves in miscible displacements in radial porous media, assuming that the density of the fluids are constant. They derived the dispersion relations as functions of the Péclet number and the viscosity ratio M , and reported a number of interesting results. For example, in contrast to the constant algebraic growth rates of purely azimuthal perturbations analyzed by Tan and Homsy (1987), axial perturbations were seen to grow with a time-dependent growth rate. Thus, there exists a critical time up to which the most dangerous axial wavenumbers are larger, but beyond which, the most dangerous azimuthal wavenumbers have higher values. The maximum growth rate of axial perturbations as well as their most dangerous and cutoff wavenumbers were found to increase with the Péclet number and the viscosity ratio M . As the Pe increased, the most dangerous wavenumber shifted towards the lower end of the spectrum. With increasing M , the most dangerous wavenumber first moved towards the lower part of the spectrum but, at later times, shifted towards the higher end.

Riaz and Meiburg (2004) carried out high-resolution 3D numerical simulations of miscible displacements with gravity override. Both homogeneous and heterogeneous porous media were considered for the quarter five-spot configuration, and the influence of viscous and gravitational effects on the overall displacement dynamics was studied. It was shown that in homogeneous porous media, the difference between the fluids' densities influences the flow, primarily by establishing a narrow gravity layer in which the effective Péclet number is enhanced due to the higher flow rate. The same effect is, however, suppressed to some extent in heterogeneous porous media due to the coupling between the viscous and permeability vorticity fields. Their simulations also indicated that when the viscous wavelength is much larger than the permeability wavelength, gravity override becomes more effective because the same coupling is less pronounced.

The effect of density variations on the stability of miscible displacements was studied by Riaz *et al.* (2006). They carried out a linear stability analysis of miscible flows in porous media driven by density variations. An important application of such a problem is carbon dioxide sequestration in saline aquifers, the dissolution of which in the underlying brine leads to a local density increase that results in a gravitational instability. They found, among other results, that for large times, both the maximum growth rate and the most dangerous mode decay as $t^{1/4}$. The long-wave and the short-wave cutoff modes scale as $t^{1/5}$ and $t^{4/5}$, respectively. High-resolution numerical simulations at short times yielded results that were in good agreement with the linear stability predictions. At later times, macroscopic fingers displayed intense nonlinear interactions that significantly influenced both the front propagation speed and the overall mixing rate.

13.9

Stochastic Models

In addition to the traditional models that we described, several models of miscible displacement processes in porous media have been developed for both the laboratory and field-scale porous media in which probabilistic concepts were utilized.

13.9.1

Diffusion-Limited Aggregation

In the diffusion-limited aggregation (DLA) model, one starts with an occupied site – the *seed* – of a lattice, located either at the center of the lattice or on one of its edges. Random walkers are released one at a time, far from the seed particle and are allowed to move randomly on the lattice (for random walk simulation see Chapters 9 and 10). If they visit an empty site adjacent (nearest-neighbor) to an occupied one, the aggregate of the occupied sites advances by one site and occupies the last site visited by the walker. The walker is removed, another one is released and so on. After a large number of particles have joined the growing cluster, the aggregate takes on a random structure with many branches, an example of which is shown in Figure 13.9.

To see the relation with miscible displacements, imagine that the aggregate represents a displacing fluid, and the empty sites represent the displaced or the defending fluid. Thus, the original seed particle represents the point at which the displacing fluid is injected into the system. Since the particles perform their random walks on the empty sites, the probability $P(\mathbf{r})$ of finding the particle at a position $\mathbf{r}$ in this region satisfies the Laplace's equation, $\nabla^2 P = 0$. Because the walkers never move into the aggregate itself, the probability of finding them there is zero, $P = 0$. If the walkers are reflected at the “walls” of the system, one has $(\nabla P)_n = 0$, where n denotes normal to the wall. Finally, the mean speed at which the front between the displacing and displaced fluids (between the aggregate and the empty sites) advances is proportional to the probability on the side of the displaced fluid next to

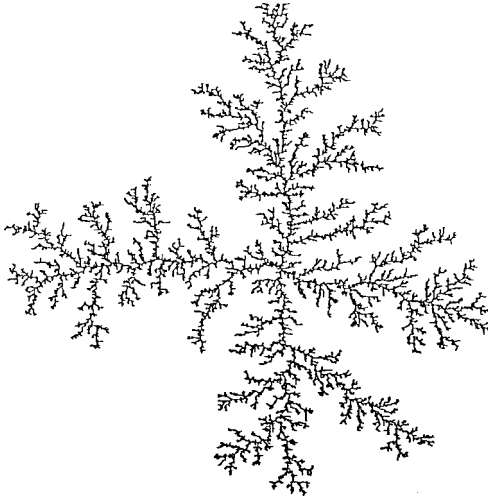


Figure 13.9 A typical two-dimensional diffusion-limited aggregate.

the front, that is, $\mathbf{v} = (\nabla P)_n$. However, these are essentially the same equations for the displacement of a viscous fluid by a miscible inviscid one (one with zero viscosity) *in the absence of dispersion*.

The one-to-one correspondence can be understood by realizing that in limit of an inviscid displacing fluid, the continuity equation, $\nabla \cdot \mathbf{v} = 0$, and the Darcy's law, $\mathbf{v} \propto \nabla P$, yield $\nabla^2 P = 0$ for the displaced fluid, and $P \simeq \text{constant}$ for the displacing fluid. However, because in the absence of dispersion the problem is linear, the constant is arbitrary and, thus, it is taken to be zero. In other words, a miscible displacement in the limits $M = \infty$ (where M is now the ratio of the viscosities of the displaced and displacing fluids) and no dispersion is simulated by the DLA algorithm. The DLA model was originally proposed by Witten and Sander (1981) for simulating aggregation of small particles. Meakin and many others studied it extensively in the 1980s and 1990s (see Meakin, 1998, for a review of the subject). Paterson (1984) was the first to point out the link between the DLA model and miscible displacements. Bogoyavlenskiy (2001) proved the existence of a one-to-one correspondence between a mean-field DLA model and the Hill–Saffman–Taylor instability problem in a Hele-Shaw cell (but not necessarily a porous medium).

If in an unstable miscible displacement, we reverse the direction of the pressure gradient and allow the more viscous fluid to displace the less viscous (or inviscid) one, the displacement will be stable. This process can be simulated by allowing the displaced fluid to advance each time n_V visits occur. Such an algorithm was suggested by Paterson (1984) and Tang (1985), and may be called the *anti-DLA* since it essentially represents the reverse of the DLA process. Paterson (1984) compared the results of such simulations with the experimental data of Habermann (1960) and found reasonable agreement. Tang (1985) compared his results with the ex-

act steady-state solution of Saffman and Taylor (1958), and the exact unsteady-state solution of Shraiman and Bensimon (1984), and found excellent agreement.

There is, however, a fundamental problem with the analogy between the DLA model and miscible displacements. Recall that a miscible displacement may be unstable if $M > 1$. The DLA algorithm produces an aggregate that (1) has a fractal structure with a fractal dimension, $D_f \simeq 1.7$ and 2.45 , in 2D and 3D, respectively, and (2) contains a large number of very tiny fingers in which tip splitting occurs at all times (see Figure 13.9). However, experiments indicate otherwise. Chen and Wilkinson (1985) displaced glycerine by oil in an etched-glass network and showed that if the system is perfectly homogeneous – if all the pores have the same radius – then the fingers form ordered (dendrites) patterns in which growth occurs mostly along the coordinates of the network, whereas the DLA model still generates a random fractal for exactly the same situation. The reason is that the surface of a DLA structure is dominated by noise: random walkers taking *random trajectories* that start far from the surface of the aggregate arrive at the vicinity of the surface one at a time. The random trajectories make the surface very rough. Thus, one must somehow reduce the noise and randomness of these trajectories. Two algorithms were introduced to accomplish this.

In one algorithm, one introduces a *sticking probability* p_s , the probability that the front or the aggregate advances by one unit once a random walker is in an empty site next to it. The DLA case corresponds to $p_s = 1$ because as soon as a random walker arrives in the vicinity of the aggregate, it joins it. If we allow p_s to be very small, however, the surface of the aggregate becomes very smooth because a random walker will encounter the front many times (on average roughly $1/p_s$ times) before the front actually advances. Such an algorithm was developed by Meakin (1986) and does result in a smooth surface. In the second algorithm, which was proposed by Tang (1985) and Szép *et al.* (1985), each time an empty site next to the front is visited by a random walker, a counter registers the event. The front does not advance to an empty site in its neighborhood unless it has been visited at least n_v times, so that $n_v = 1$ corresponds to the DLA case, and $n_v \rightarrow \infty$ represents the noiseless limit of the DLA model. The two algorithms are, in fact, equivalent. Kertész and Vicsek (1986) showed that such modification of the DLA algorithm can reproduce the patterns obtained by Chen and Wilkinson (see also Siddiqui and Sahimi, 1990a).

Several other aspects of DLA-based simulation of miscible displacements deserve careful considerations.

1. The first question that arises is whether this type of simulation can be generalized to the case of finite values of M . Recall that the DLA model corresponds to $M = \infty$.
2. The second question is whether this type of simulation can be a *quantitative* tool for simulating miscible displacements in a *disordered* porous medium, at least in the limit $M = \infty$.
3. One should recognize the fact that this type of simulation exhibits a sensitive dependence on the lattice size and, therefore, it is highly important to establish

- links between the parameters of the simulations and the physical parameters that can be measured.
4. One needs to generalize the DLA model to be used for simulating miscible displacements in a porous medium with a pore size distribution, or for one with a permeability distribution (as in a large-scale porous medium).
 5. Another important question is whether it is possible to generalize a DLA-based model to include the effect of dispersion.

Before closing this section, we should mention that the DLA algorithm was also generalized to include the effect of surface tension that is suitable for an immiscible displacement in a Hele-Shaw cell, the classical Hill–Saffman–Taylor problem described earlier. The generalization model was suggested by Szép *et al.* (1985) and Kadanoff (1985). The main physical fact to include in the model is to force the probability $P(r)$ at the interface between the two fluids to take on the form

$$P(r) = a_1\kappa + a_2, \quad (13.47)$$

where κ is the curvature of the interface, a_1 is proportional to the surface tension, and a_2 is a constant. Thus, one allows the walkers to leave the interface and walk through the displacing fluid until they finally reach the interface again. The walkers are allowed to leave the displacing fluid with a probability proportional to $P(r)$ given by Eq. (13.48), which measures the net flux of walkers through each interface bond, and then moves the displaced or displacing fluid forward whenever the flux is $-n_V$ or $+n_V$. In this manner, one has a meaningful model for simulating *immiscible* displacements in a Hele-Shaw cell.

Various versions of this basic model were used by Sahimi and Yortsos (1985), Meakin *et al.* (1987), Sarkar and Jensen (1987), and Tao *et al.* (1988) to investigate different aspects of the problem. Vicsek (1984) suggested a variant of the DLA in which the particles stick to the aggregate with a probability proportional to $P(r)$ given by Eq. (13.48), *without* the reshuffling of the interface suggested by Kadanoff (1985) and Szép *et al.* (1985). Sarkar (1985) showed that Vicsek's model simulates the *early stages* of the Hill–Saffman–Taylor phenomenon. Finally, Liang (1986) successfully applied the Kadanoff–Szép *et al.* model to the study of immiscible displacements in a Hele-Shaw cell. He compared his results with those obtained by other methods (see Bensimon *et al.*, 1986, for a review) and the experimental data, and found good agreement.

13.9.2

The Dielectric Breakdown Model

The dielectric breakdown (DB) model was proposed by Niemeyer *et al.* (1984) to study dielectric breakdown phenomena in disordered solids, but it can also be used for simulating viscous fingers in the absence of dispersion because in the absence of dispersion, the governing equations for the two phenomena are similar. In the DB algorithm, a discrete version of the Laplace's equation is solved. This means, for example, that on a square lattice, the unknown function F_i at lattice site i is

given by $4F_i = F_{i+1} + F_{i-1} + F_{i+L} + F_{i-L}$, where L is the linear size of the lattice, and $(i+1, i-1, i+L, i-L)$ are the nearest-neighbor sites of i . Similar equations are used for other lattice topologies. For example, for the simple-cubic lattice, one has $6F_i = F_{i+1} + F_{i-1} + F_{i+L} + F_{i-L} + F_{i+L^2} + F_{i-L^2}$. As the boundary condition, the unknown function is specified at the boundary. In the context of a miscible displacement, the unknown function is the pressure, so the simulation is appropriate only for the case of a viscous fluid displaced by an inviscid one ($M = \infty$). At each step of the simulation, one site on the boundary between the two regions (between the displaced and displacing fluids) is selected for advancement of the front between the two regions, with the probability of selection being proportional to a power of the gradient of the unknown function, for example, the pressure in the miscible displacement problem. The same model was used by Ben-Jacob *et al.* (1985) to explain dense branching morphology formed in various fluid displacements.

The DB model can also be simulated by a random walk technique. The method would be similar to that of DLA, except that in the DB model, the front advances to a nearest-neighbor empty site if this site is visited by a random walker *that also crosses the front*. However, in the DLA model, the front advances as soon as the empty site is visited by the walker. Thus, the boundary conditions at the front are *not* the same for the DB and DLA models. However, although the difference between the DLA and DM models may seem minor, the results are very different, indicating the sensitivity of the two models to the boundary conditions.

13.9.3

The Gradient-Governed Growth Model

DeGregoria (1985) and Sherwood and Nittmann (1986) introduced an algorithm for simulating miscible displacements for finite values of M , and in the absence of dispersion. The model is usually called the gradient-governed growth (GGG) model, and is essentially an extension of the DB model to the case when both fluids have a finite viscosity. Similar to the DB model, the discrete version of the Laplace's equation is solved in the region occupied by each fluid to yield the pressure field. The front between the two fluids is advanced at each time step with the selection probability being proportional to the local pressure gradient between a point on the front and the point to which the front is to advance. The model is *wrong* in the (pore-scale) microscopic sense because the velocity field is determined assuming the entire front is moving instantaneously, yet only one bond at the front is moved at a time.

DeGregoria (1985) and Sherwood and Nittmann (1986) used the GGG model to simulate miscible displacement for finite values of M in the absence of dispersion. Using a 100×100 square network, he obtained reasonable agreement with Habermann (1960)'s data. Sherwood (1986) used the same model for investigating the size distribution of the islands of the displaced fluid that are formed when the displacing fluid completely surrounds a portion of the displaced fluid.

13.9.4

The Two-Walkers Model

This model was proposed by Siddiqui and Sahimi (1990a) for simulating miscible displacements at finite values of M in the absence of dispersion *using random walkers only*. Since the pressure in each fluid region satisfies the Laplace's equation, one random walker for each fluid region (displacing and displaced) is used. Because in the absence of dispersion and surface tension the front always advances forward, both random walkers advance the front, upon contacting it, with a probability proportional to $p_1 = (M + 1)^{-1}$ for the particle in the displacing fluid region and $p_2 = M/(M + 1)$ for the particle in the displaced fluid region. As such, the model can be thought of as the random walk version of the GGG model. Due to different mobilities of the two fluids, the lengths of each step of the random walkers are *different* in each fluid region.

The results obtained with this model are in complete agreement with those obtained with DB and GGG models as well as with the experimental expectations. Figure 13.10 presents samples of the patterns obtained with this model with the triangular lattice. Note the dendritic structure of the aggregate for $M = 1000$, in agreement with the experiments of Chen and Wilkinson (1985) that indicate growth of the cluster along the main symmetry axes of the lattice, and the compact structure of the aggregate for $M = 5$, which is what is expected for a displacement process in which M is close to unity. Siddiqui and Sahimi (1990a) also generalized the model to the case in which there is a pore size or pore permeability distribution. In this case, the random walkers take each step with a probability proportional to the (pore or region) permeabilities. A somewhat similar model was proposed by Leclerc and Neale (1988).

Among the discrete models described so far, the DLA model and its two-walker generalization are the only ones that *only* use random walkers and, thus, from a computational point of view, are very efficient. The relevant question is, therefore, whether such random walk models can provide quantitative predictions for misci-

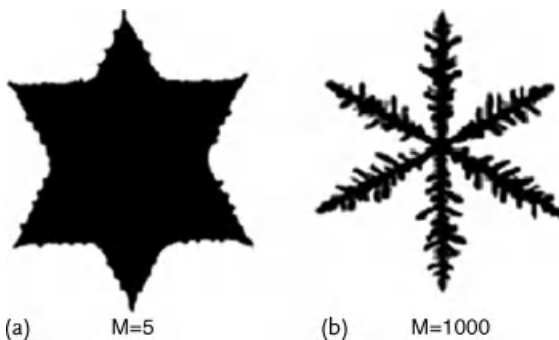


Figure 13.10 Patterns of displacements obtained with the two-walkers model on the triangular network for two values of the viscosity ratio M where (a) $M = 5$ and (b) $M = 1000$. Note the rough surface of the pattern for $M = 5$ (after Siddiqui and Sahimi, 1990b).

ble displacements in which dispersion plays no important role. At first, the answer may seem affirmative since the governing equations for both phenomena are identical. However, this only guarantees that the *universal* properties of the two processes, such as, the fractal dimension that characterizes the structure of the displacing fluid cluster, to be the same, but not necessarily the equality of nonuniversal quantities of interest. For example, as far as a petroleum or chemical engineer is concerned, the most important quantity to predict is the volume fraction of the displaced fluid at the breakthrough point, that is, the point at which the displacing fluid reaches the production point (well) since this is a quantitative measure of the efficiency of the displacement. As explained by Sahimi and Siddiqui (1987), the proper comparison is between the DLA-like models that only use random walkers and a deterministic model in the absence of dispersion because in an actual experiment, the front does not advance one pore at a time, as in the DLA, DB and GGG models and their generalizations, but advances in several pores simultaneously, which is the basis of a deterministic model that will be described shortly.

13.9.5

Stochastic Models with Dispersion Included

Two stochastic models that take into account the effect of physical dispersion were developed about two decades ago, the predictions of which agree quantitatively with the relevant experimental data. One model is due to King and Scher (1987, 1990). Consider, first, the case of miscible displacements *without* dispersion. For point injection of fluids, the governing equations are

$$\frac{\partial C}{\partial t} + \mathbf{u} \cdot \nabla C = \delta^2(\mathbf{x}) , \quad (13.48)$$

$$\mathbf{u} = \frac{\mathbf{v}}{\hat{Q}} = \nabla \times (\psi \hat{\mathbf{z}}) , \quad (13.49)$$

where $\hat{Q}$ is the injection rate (volume per unit thickness per unit time), ψ is the stream function, and $\delta^2(\mathbf{x})$ is the 2D Dirac delta function. The injected volume of fluid provides a natural time variable

$$\tau = \frac{\int_0^t \hat{Q}(t') dt'}{\psi} . \quad (13.50)$$

In the absence of dispersion, the solution is simple: A concentration bank $C = 1$ (i.e., the pure fluid) displacing $C = 0$. In general, however, the concentrations shall not form a bank since dispersion intervenes and develops a mixed zone. To develop a probabilistic model that takes this effect into account, King and Scher (1987, 1990) interpreted $(\partial C / \partial \tau) d^2 x$ as a 2D probability density function for the concentration evolution. According to Eqs. (13.49) and (13.50), one has

$$\frac{\partial C}{\partial \tau} d^2 x = - \frac{\partial C}{\partial \xi_1} \frac{\partial \psi}{\partial \xi_2} d\xi_1 d\xi_2 = -dC d\psi , \quad (13.51)$$

where ξ_1 and ξ_2 are local tangential and normal coordinates on the front. Equation (13.52) is now given a probabilistic interpretation: The probability of concentration evolution at $\mathbf{x}$ (i.e., $(\partial C/\partial \tau) d^2 \mathbf{x}$) is the product of the probability $d\psi$ of fluid flow through $\mathbf{x}$ and the probability dC of a concentration gradient moving through $\mathbf{x}$. For a given *realization* of the porous medium, one samples the cumulants C and ψ , that is, determines the flux contour $C = r_1$ and the streamline $\psi = r_2$, and calculates their intersection in the plane, which is also the point at which concentration is modified, that is, where r_1 and r_2 are two random numbers uniformly distributed in $(0, 1)$.

In such a simulation, one must employ a probabilistic interpretation of the FD version of Eq. (13.49). We integrate Eq. (13.49) over a rectangular spatial region A_{ij} and time interval $\Delta \tau$ to obtain δC_{ij} , the change in the average concentration C_{ij} which is given by

$$\delta C_{ij} = -\frac{\Delta \tau}{\Delta x \Delta y} \oint_{\partial A_{ij}} \bar{C} d\psi. \quad (13.52)$$

Obviously, if $\delta C_{ij}/\Delta C$ is properly normalized, then it can be interpreted as the growth probability at site ij (i.e., as the probability that the displacing fluid and the front between the two fluids advance). One must then fix ΔC . If we set $\Delta C = 1$, we would have a situation similar to the DLA model, that is, a cluster of fluid at $C = 1$ representing the aggregate of the displacing fluid, facing a cluster of $C = 0$ representing the displaced fluid. According to Eq. (13.53), the growth probability is nonzero only when the boundary integral overlaps the edge of the cluster of the displacing fluid mixed with the displaced fluid. The algorithm has the advantage that finite values of the viscosity ratio can be used in the simulations. However, simulating very large systems based on such an algorithm is excessively time consuming.

The effect of dispersion can be added to the model. Consider the static case, $\mathbf{v} = 0$. The evolution equation is simply the diffusion equation, $\partial C/\partial T - D_L \nabla^2 C = 0$, where $T = D_L t$ (King and Scher assumed that, $D_L = D_T$, which is, however, not justified). In discrete form,

$$\delta C_{ij} = \frac{\Delta T}{\Delta x \Delta y} \oint_{\partial A_{ij}} \hat{\mathbf{n}} \cdot \nabla \bar{C} dl, \quad (13.53)$$

which should be compared with Eq. (13.53). If the time step ΔT satisfies $\Delta T/(\Delta x \Delta y) \leq \Delta C$, then

$$\frac{\delta C_{ij}}{\Delta C} = -\sum_{\text{faces}} \oint_{\partial A_{ij}} \left(\frac{\Delta T}{\Delta x \Delta y \Delta C} \right) (-\hat{\mathbf{n}} \cdot \nabla \bar{C}) dl. \quad (13.54)$$

For the complete problem – convection + dispersion – we split the time evolution as

$$\phi \frac{\partial C}{\partial t} = \hat{Q} \frac{\partial C}{\partial \tau} + \phi D_L \frac{\partial C}{\partial T}. \quad (13.55)$$

In Eq. (13.56), $\partial/\partial t$ and $\partial/\partial T$ represent separate convection and dispersion processes. Convective evolution is described by Eq. (53), while the dispersion evolution is represented by Eq. (13.56). Thus, we have a sequential finger evolution in which convection initiates the growth, which is then modified or moderated by dispersion. If we fix the time interval δt , then $\delta \tau = (\hat{Q}/\psi)\delta t$, and $\delta T = D_1 \delta t$, which then define the Péclet number, $Pe = \delta \tau / \delta T$. In practice, $\delta \tau$ is set by $\delta \tau = \Delta \tau = \Delta x \Delta y \Delta C$, implying that $\Delta T = \Delta \tau / Pe$. In most practical cases, $Pe > 1$, and Eq. (13.56) is properly normalized. However, if $Pe < 1$, then δT is subdivided into n_D intervals to obtain $\Delta T = (\delta \tau / Pe) n_D$, where $n_D > Pe^{-1}$. King and Scher (1990) simulated miscible displacements, with dispersion effect included, for various values of the viscosity ratio and obtained reasonable agreement with the experimental data.

The second probabilistic model is due to Araktingi and Orr (1988). In their model, the porous medium is represented by a 3D (2D) grid of cubic (square) blocks. At the beginning of each time step, the pressure field is calculated given the distribution of the permeability and the fluid viscosities. Tracer particles that carry a finite concentration of the displacing fluid are injected into the porous medium and are moved with velocities based on the pressure field. The velocities are calculated at the midpoint between grid nodes. Velocities for particles that are not on such nodes are obtained by linear interpolation. After moving the particles by convection to their current positions, the effect of dispersion is simulated by random perturbations of particle positions in the longitudinal and transverse directions. Since for diffusive dispersion the standard deviations of the particles' motion are

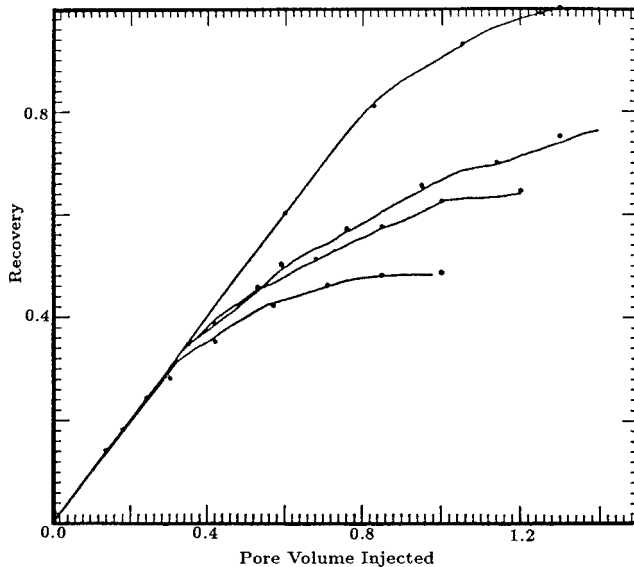


Figure 13.11 Comparison of the predictions of the Araktingi–Orr model (curves) with the experimental data (symbols) of Blackwell *et al.* (1959). The results are, from top to bottom, for $M = 5, 86, 150$, and 375 (after Araktingi and Orr, 1988).

given by $\sigma_x = \sqrt{2D_L t}$ and $\sigma_y = \sqrt{2D_T t}$, the distribution of the particles about a mean position is simulated by multiplying the standard deviations by a number between -6 and $+6$. This number is obtained by adding a sequence of 12 random numbers, distributed normally with a zero mean and unit standard deviation to -6 . The values $+6$ and -6 are used because, on a practical basis, the probability of a particle moving beyond six standard deviations on either side of the mean is less than 1%. After the particles arrive at their new positions, the current time step is determined. To avoid having particles travel a distance greater than a grid block, the time step is chosen to allow movement equal to half of a grid block length (or width), traveled at the highest existing velocity. The new pressure field is calculated, and a new position for each particle is determined. The procedure is repeated many times. Araktingi and Orr (1988) compared their results with the experimental data of Blackwell *et al.* (1959). Figure 13.11 shows the comparison; the agreement is very good. Generalization of the model to irregular computational grids was achieved by Ebrahimi and Sahimi (2004).

13.10

Pore Network Models

Pore network models were developed for investigating viscous fingers in physical pore networks. Nobles and Janzen (1958) had already used analog resistor networks to study the effect of viscosity ratio M on miscible displacements. Random networks and deterministic flow models were originally proposed by Simon and Kelsey (1971, 1972), but their model was too simple and the networks used were too small. There are two pore network models, one of which is applicable to miscible displacements in the absence of dispersion, whereas the second one takes into account the effect of dispersion.

Let us first describe the case in which dispersion is neglected. The porous medium is represented by a network of interconnected pores, usually assumed to be cylindrical pores with distributed radii. Consider a pore of length l_{ij} and radius R_{ij} that connects nodes i and j , of which a portion x_{ij} is occupied by the displacing fluid and the rest by the displaced fluid. The pressure difference $P_i - P_j$ along the tube pore given by

$$P_i - P_j = \frac{8\mu_2 \left(\frac{x_{ij}}{M} + l_{ij} - x_{ij} \right) Q_{ij}}{\pi R_{ij}^4} = \frac{Q_{ij}}{g_{ij}}, \quad (13.56)$$

where μ_2 is the viscosity of the displaced fluid, Q_{ij} is the flow rate in the pore between i and j , and g_{ij} is the hydraulic conductance of the pore. At each node i of the network, we have conservation of fluid fluxes, $\sum_j Q_{ij} = 0$, which, when written for every interior node of the network, yields a set of linear equations for the nodal pressures. The boundary conditions are that at the injection point, $P = 1$, while at the production point $P = 0$. After determining the nodal pressures, the front

between the two fluids is moved a distance

$$\Delta x_{ij} = \frac{Q_{ij}}{\pi R_{ij}^2} \Delta t, \quad (13.57)$$

into one of the pores adjacent to the interface. The time step Δt is selected to be the time necessary to exactly move the front to reach a node through the fastest pore. All other front's positions in the pores are then updated (i.e., they are moved into the slower pores adjacent to the front to *partially* fill them up), and for the new configurations of the displacing and displaced fluids, calculate the pressure field and repeat the entire process.

The model and its variations were used by Chen and Wilkinson (1985), King (1987), Siddiqui and Sahimi (1990a,b), Ferer *et al.* (1992), and Tian and Yao (2001) to study miscible displacements without dispersion. The main advantage of the method is that it allows one to investigate the effect of pore- or large-scale heterogeneities on the displacement process. It is also free of the type of noise that is generated by the DLA-like models. Figure 13.12 compares the predictions of the model with Habermann's data, and the agreement is good.

As discussed above, an important question is regarding such nonuniversal properties of miscible displacements as their sweep efficiency can be predicted by DLA-like models. Answering this question requires a careful comparison between the predictions of the pore network model in the absence of dispersion and those of DLA-like models. Chan *et al.* (1986, 1988), Sahimi and Siddiqui (1987), and Siddiqui and Sahimi (1990a) studied this problem in detail. Sahimi and Siddiqui (1987) computed the sweep efficiency using both models and found them to be generally different. Chan *et al.* (1986, 1988) argued essentially along the same lines, except that their model of pore space was different from that of Sahimi and Siddiqui. In

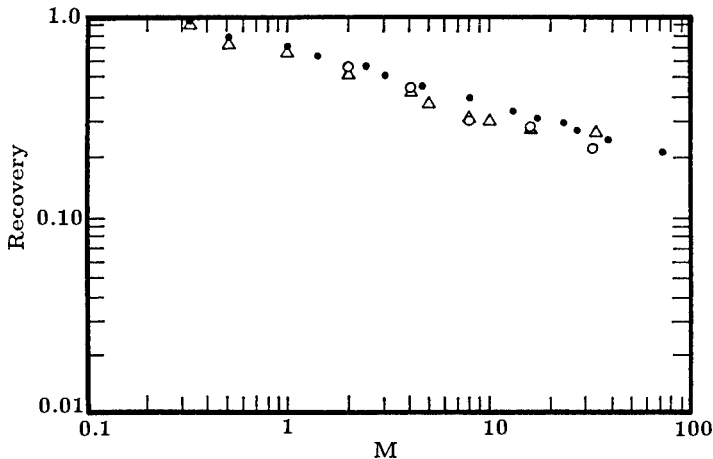


Figure 13.12 Comparison of the predictions of the two-walkers model (triangles) and the pore network model (open circles) with the experimental data of Habermann (solid circles) (after Siddiqui and Sahimi, 1990a).

their model, the porous medium was represented by a network of interconnected *pores* and *chambers* (pore bodies). The pores have a small diameter and, thus, contribute most of the resistance to fluid flow. The pores connect the grid points of the network a distance l apart at which the chambers are located. The chambers have volumes much greater than those of the pores, thus making a negligible contribution to the hydrodynamic resistance. The fluid capacity of each chamber, that is, its volume per specified volume l^3 , is randomly distributed. Chan *et al.* showed that, unless the distribution of the fluid capacity of the chambers is exponential, the predicted values of the sweep efficiency by their model and the DLA model will be different. The conclusion is that although the DLA-like models often provide a good description of the universal properties of miscible displacements without dispersion, the mapping between the two problems is not exact but approximate, although a very good one in many situations.

The second deterministic network model is capable of taking into account the effect of dispersion. As in the first model, we first calculate the pressure field throughout the network. There is, however, one difference between this case and the first model: the portion x_{ij} is occupied by a *mixture* of displacing and displaced fluid with an effective viscosity μ_m given by, for example, Eq. (13.1). Thus, M should be calculated and used in Eq. (13.57) based on the effective viscosity μ_m and the viscosity μ_2 of the displaced fluid, that is, $M = \mu_2/\mu_m$. One then assumes that within each pore, a 1D CD equation governs the concentration distribution C . Using the fact that the net mass flux reaching any node j is zero, $\sum_j S_{ij} J_{ij} = 0$, where $J_{ij} = v_{ij} C_i - D_m \partial C_i / \partial x$ is the total flux (convective + diffusive) in the pore that connects sites i and j , and S_{ij} is the cross-section area of pore ij . Also, by employing the distribution of the pore fluid velocities, one determines the concentration distribution in the entire network by writing down this equation for every node of the network. This yields a set of equations for the nodal concentrations C'_j and is solved numerically. Once the concentration distribution is determined, one proceeds as in the first case, except that the distance Δx_{ij} that the front moves in a pore is given by $\Delta x_{ij} = J_{ij} \Delta t / (\pi R_{ij}^2)$.

13.11

Crossover from Fractal to Compact Displacement

Since the universal properties of viscous fingering in the limit $M \rightarrow \infty$ and without dispersion, and those of the DLA model are the same, one may conclude that viscous fingers are fractal objects in the limit $M = \infty$. However, what about the case of viscous fingers at finite values of M ? We already know that viscous fingers may be unstable if $M > 1$, which might mean that they are fractal objects for *any* $M > 1$, with the instability manifesting itself in the fractal structure. This would imply, however, that the density of the displacing fluid in the region that it occupies would vanish as the displacement proceeds, as this is a general property of any fractal object. This also means that very thin sections of fluids would have to support a vast and tenuous network of the fingers at the tip.

In fact, if the displaced fluid has a finite mobility, one would expect the fingers to become *thicker*, as opposed to the thinner fingers with the fractal structure. However, for $M > 1$, the displacement can be unstable if dispersion effects are absent, and we cannot expect it to be smooth. The cluster of the displacing fluid is also not compact since there is no intermediate length scale between the size of the system and that of a pore. Thus, as argued by King (1987), only the *surface* of the viscous fingers – the front between the two fluids – can have a fractal-like character, which can be interpreted as the manifestation of the instability of the process. This argument is supported by the simulations of King (1987), Blunt and King (1988), Siddiqui and Sahimi (1990a), and Ferer *et al.* (1992) who used the pore network model, and by those of Siddiqui and Sahimi (1990b) who used the two-walkers method.

Even the surface roughness and its fractal character should gradually disappear as the size of the system increases. This was demonstrated by King and Scher (1990) who argued that if the linear size of the system exceeds a crossover size L_{co} , then there would be a crossover from a fractal-like behavior to a compact displacement, where L_{co} is given by

$$L_{co} \sim \left[\frac{1}{2} (M - 1) \right]^{\frac{1}{D_f}}, \quad (13.58)$$

where D_f is the fractal dimension of DLA aggregates. King and Scher (1990) called this phenomenon *viscous relaxation*. Earlier Monte Carlo simulations of DeGregoria (1986) using the GGG model had already provided evidence for such a phenomenon. DeGregoria observed that, for finite values of M , the volume fraction of displaced fluid increases as the size of the system increases, which implies increased stability. Note that Eq. (13.59) implies that no such crossover occurs if $M \rightarrow \infty$.

The crossover between fractal and nonfractal miscible displacements has two practical implications. The first is that the scale up of numerical or laboratory experiments of unstable displacements ought to be done with caution since, as suggested by Eq. (13.59), there will always be a crossover to a stable displacement if one waits long enough, or if the scale-up is done for a large enough scale. The second implication is that the heterogeneities on a scale of the order of the crossover length L_{co} or larger dominate viscous fingering.

13.12

Miscible Displacements in Large-Scale Porous Media

Miscible displacements in large-scale (LS) porous media – those in which the spatial distribution of the permeability is very broad – have also been studied by various methods, although such studies are not as extensive as those for laboratory-scale porous media. For example, Tan and Homsy (1992) used a continuum model to study miscible displacements in the LS porous media in which the heterogeneities

were modeled as stationary random functions of space. The permeability correlation length ξ_K was finite. They found that the fingers grow linearly in time in a manner similar to that of laboratory-scale porous media described earlier, which is not surprising because ξ_K was finite.

Paterson (1987) modified the DLA-algorithm to account for the effect of the spatial distribution of the permeabilities. A notable example of discrete simulation of miscible displacements in the LS porous media in which the effects of dispersion, mobility, and spatial distribution of the permeability are taken into account is the work of Araktingi and Orr (1988) and Tchelepi and Orr (1993, 1994). In their work, the mean of the permeability distribution was independent of the location (first-order stationarity), and the spatial correlation between any two regions depended only on the distance between them. A heterogeneity index H_i was used to characterize a permeability field defined by

$$H_i = \sigma_{\ln K}^2 \hat{\xi}_K, \quad (13.59)$$

where $\sigma_{\ln K}^2$ is the variance of log permeability, and $\hat{\xi}_K$ is the dimensionless correlation length of permeability, $\hat{\xi}_K = \xi_K/L$, with L being the porous medium's length. Note that such a definition of H_i is somewhat different from that proposed by Koval. As discussed earlier, H_i combines the variability (as measured by $\sigma_{\ln K}^2$) and the spatial correlations of the permeability field.

Figure 13.13 compares the simulation results for two values of M and two values of H_i . It is clear that for low values of H_i , that is, a more homogeneous porous medium, the effect of M is strong, which is expected. However, as H_i increases, the effect of M diminishes, and the shape of the fingers is dominated by the permeability heterogeneities. For example, for $H_i = 0.77$, the shapes of the fingers are almost identical for $M = 1$ and 20. If this is the case, then a simple general-

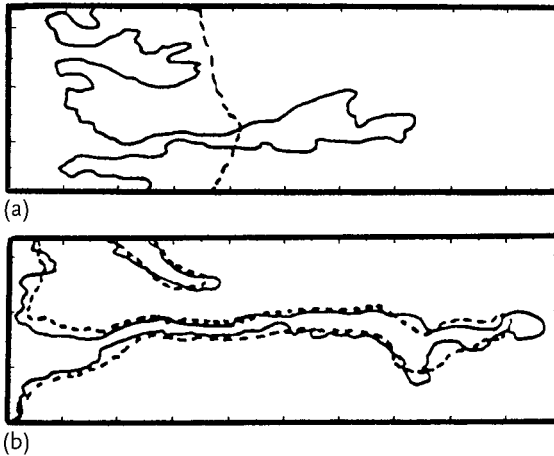


Figure 13.13 Comparison of the shapes of viscous fingers for the heterogeneity index $H_i = 0.25$ (a) and $H_i = 0.77$ (b), and viscosity ratio $M = 1$ (dashed curves) and $M = 20$ (solid curves) (after Araktingi and Orr, 1988).

ization of a DLA-like model (or any other random-walk method) should suffice for simulating miscible displacements in the LS porous media because as discussed in Chapter 11, pore-level dispersion is not important in the LS porous media, and the fluctuations of the velocity field, induced by the permeability field, is the most important factor. Such an effect is easily captured by a DLA-like model.

One of the factors that distinguishes a LS porous medium from a laboratory-scale one is the extent of the spatial correlations in the permeability distribution. If there are no spatial correlations, or they are of finite extent, we do not expect a miscible displacement in the LS medium to be very different from that in the laboratory-scale one. One way of incorporating the effect of long-range permeability correlations is, of course, through the use of fractal statistics that we already described in Chapter 5. Emanuel *et al.* (1989) and Mathews *et al.* (1989) used such statistics in their simulations of displacement processes. They showed that using such statistics leads to significant improvement in the predictive ability of their models. They used the FD techniques for solving the governing equations, which may limit the size of the system and the extent of the correlations that can be simulated, as one usually needs a very fine grid structure with the finite-difference techniques in order to achieve reliable accuracy.

On the other hand, Sahimi and Knackstedt (1994) used a random-walk model to study miscible displacements in the LS porous media in which the permeabilities followed the statistics of fractional Brownian motion described in Chapter 5. They showed, in agreement with the work of Araktingi and Orr (1988), that M does not play any important role in the efficiency of the miscible displacement and the shape of the clusters formed, and that it is the permeability distribution that controls the efficiency of the displacement.

13.13

Miscible Displacements in Fractures

Unlike Hele-Shaw cells and disordered porous media, miscible displacements have not been studied extensively in fractures and fractured porous media. Most of such studies have used the double-porosity model of fractured porous media that was described in Chapter 12. Thus, the same type of strength and weaknesses of the model described in Chapter 12 could be expected when using the double-porosity model to study miscible displacements.

Trivedi and Babadagli (2008) studied miscible displacements in fractured porous media using scaling analysis. The goal was to identify the dimensionless groups that control the displacements. Er and Babadagli (2010) reported on an experimental study of miscible displacements in a fractured sample, focusing on the matrix-fracture interactions. They reported that the key parameters were the molecular diffusivity D_m and the longitudinal and transverse dispersion coefficients, D_L and D_T . It was necessary to attribute distinct dispersivities to the fractures and the blocks that represent the matrix.

There have been a few studies of miscible displacements in a single fracture with a rough self-affine internal surface of the type described in Chapter 12. Auradou *et al.* (2001) studied the problem experimentally in a granite fracture with rough self-affine surface. They found that for a purely normal relative displacements between the two surfaces, the displacement front is globally smooth due to the constant local distance between the surfaces. For a finite lateral displacement, however, the displacement front was found to be rough and self-affine. Drazer *et al.* (2004) studied the same problem by numerical simulations, and found that the roughness exponent of the displacement front is the same as the roughness or Hurst exponent H that characterizes the roughness of the fracture's internal surface, hence establishing a link between the dynamics of the displacement and surface morphology, which is static and fixed. The lower cutoff of the self-affinity of the displacement front was found to be dependent only on the aperture, whereas the upper cutoff grew with the lateral shift of the two surfaces, and linearly with the width of the front.

13.14

Main Considerations in Miscible Displacements

Due to their high cost, field-scale miscible displacements, particularly those that use a gas as the displacing agent, require careful design. The basic design parameters include the geology of the porous medium or reservoir; its porosity and permeability distributions; the temperature and pressure; the relative permeability curves characterizing two-phase flow in the reservoir (see Chapters 14 and 15); the amount of residual oil, and the viscosity and *minimum miscibility pressure* of the oil-in-place. The relative permeability to a fluid phase is the ratio of the permeability of that part of the pore space occupied by the fluid and the single-phase permeability of the entire pore space. The minimum miscibility pressure is typically determined phenomenologically by measuring displacement of crude oil by CO₂ (or another fluid) in a long capillary tube – usually called the *slim tube* – at a series of successively higher pressures. If we plot the amount of the displaced oil versus pressure, the resulting curve usually has a break at about 95% recovery. The pressure corresponding to that recovery is taken as the minimum miscibility pressure.

13.14.1

Reservoir Characterization and Management

In virtually all gas injection projects, the most critical decisions are made only after extensive computer simulations that attempt to optimize the amount of the fluid injected, the injection and production rates, and other operational variables. Thus, utilizing a realistic model of the reservoir is very important and, in fact, the accuracy of any predictive simulation technique is directly limited by the accuracy with which the reservoir is described. Despite the extensive sets of experimental data that are used as the input parameters, the sophistication of the simulators, the size

and speed of the computers on which the simulations are carried out, and the large number of cases that are simulated, the simulator's predictions may still be subject to very large uncertainties. Therefore, a much less expensive pilot flood is usually carried out first, and the reservoir's model is tuned by changes in the values of the input parameters in order to make the simulator's output fit the pilot data.

For the field-scale projects that have been carried out, the calculated optimal CO₂ injection volumes have ranged from 20–50% the hydrocarbon pore volume. The predicted CO₂ utilization factors range from 5 to 15 million ft³ CO₂/bbl of recovered oil. The projected ultimate oil recoveries range from 5–30% of the original oil-in-place. Such numbers only represent the consensus of the current expectations.

13.14.2

Mobility Control

As discussed earlier, the control of unfavorable mobility ratios is recognized as a major technological problem of gas-flood EOR as they produce the fingering phenomena that are a principal reason for the failure of many of the liquefied petroleum gas floods of the 1950s and early 1960s (Craig, 1970). Such problems arise because the injected gases have very small viscosities at the temperatures and pressures at which they are used, hence producing an unfavorable mobility ratio. The problems associated with the mobility are greatly aggravated, and their theoretical and experimental study greatly complicated by the facts that (1) the reservoir is highly heterogeneous at every length scale, from sub-millimeter to kilometer, and (2) the number of fluid phases is often three rather than two, that is, the miscibility is not complete. To control the mobility, many methods have been used, such as, water-alternating-gas (WAG), gas-soluble viscofiers, gelled and cross-linked polymers, and surfactants and foams. We describe one of them, namely, the WAG process.

13.14.3

Miscible Water-Alternating-Gas Process

The WAG is the only gas-flood mobility control technique that is used regularly in field applications. Its chief virtues are familiarity to the petroleum engineers as it consists of the familiar water flooding, alternated with gas flooding; simplicity, having timing and the ratio of water to gas as its only design parameters, and its low cost. It is used mostly to maintain pressure while the solvent supply is interrupted, or to stretch out injection costs by substituting water for the more expensive solvent.

Miscible WAG injection has been implemented successfully in a number of fields around the world (Christensen *et al.*, 1998). In principle, it combines the benefits of miscible gas injection and water flooding by injecting the two fluids either simultaneously or alternatively. Miscible gas injection has excellent microscopic sweep efficiency, but poor macroscopic sweep efficiency due to fingering

and gravity override that were already described. Furthermore, it is expensive to implement. The optimal ratio for simultaneous WAG injection in a relatively homogeneous reservoir can be estimated by matching the advance rates of the water-oil and solvent-oil displacement fronts. Water flooding, on the other hand, is cheaper and less vulnerable to gravity segregation and frontal instabilities. However, the residual oil saturations after water flooding are relatively high. Johns *et al.* (2003) studied optimization of the WAG processes for enriched gas floods, particularly as a primary recovery method. Al-Shuraiqi *et al.* (2003) carried out laboratory investigations of first-contact miscible WAG to study the effects of the WAG ratio and flow rate on the recovery efficiency. Their experiments (using bead-packs) indicated that (1) recovery from the WAG may vary with rate, even when gravity is neglected, and (2) water-solvent and water-oil relative permeabilities may not be the same if they are first-contact miscible.

13.14.4

Relative Permeabilities

The EOR processes that are carried out under near miscible conditions require relative permeability data to calculate the flow behavior of low interfacial tension (IFT) fluids. In systems where the displacing and displaced fluids are miscible and the flow velocity is low enough that the process is controlled by diffusion, the fractional flow versus saturation curve is a 45° line, and is referred to as miscible relative permeability function. The term *near-miscible relative permeability functions* is used to denote the curves found in the region between the immiscible limit (to be studied in Chapters 14 and 15) and miscible limits. If the IFT is finite and there is no mass transfer, the relative permeabilities of the phases are more complex (see Chapter 14).

It is not yet completely clear how near miscibility changes the relative permeabilities, and which parameters are controlling the change (Blom, 1999). Some investigators (Amaefule and Handy, 1982; Henderson *et al.*, 1996) reported that the relative permeability to the non-wetting phase is affected more strongly, while others (Asar and Handy, 1988; Schechter and Haynes, 1992) reported the same for the wetting phase. For example, Cinar and Orr (2004) reported a 10-fold increase of the non-wetting phase relative permeabilities against a 100-fold decrease in the IFT. Others did not find any effect of the IFT at all (Kalaydjian *et al.*, 1996). Equally contradictory are the reports on the effect of the flow velocity on the near-miscible relative permeabilities. Some investigators (Fulcher *et al.*, 1985) reported no effect, whereas others (Ali *et al.*, 1997; Henderson *et al.*, 2000; and references therein) observed an increase with the velocity.

There appear to be two conflicting views on the mechanism that controls the change in the relative permeability. Bardon and Longeron (1980), Jerauld (1997) and Blom *et al.* (2000) argued that a low IFT affects the relative permeabilities through the capillary number, the ratio of the viscous and capillary forces (see Chapters 4 and 14). Most of the authors suggest, however, that there is a threshold IFT below which the capillary-number dependence becomes important. On

the other hand, Haniff and Ali (1990), Munkerud (1995), and others interpreted their relative permeability data in terms of the IFT alone. In two of the cases, the interpretation was done in view of the fact that a transition from partial wetting to complete wetting, as predicted by Cahn (1977), may affect the mobility of both phases. The influence of such a wetting transition cannot be described in terms of the capillary number because it is directly induced by a change in the interfacial tension between the near-miscible phases. Blom and Hagoort (1998) reviewed the progress in resolving some of these issues.

13.14.5

Upscaling

In a miscible injection, the effects of channeling and fingering, which occur due to the heterogeneity and adverse mobility ratio between the injected fluid and oil, must be properly accounted for in order to obtain an accurate estimate of the displacement efficiency. However, the resolution of the required computational grid would be extremely high. This fact, coupled with the high number of components and complex phase behavior, render fine-scale simulations of miscible processes prohibitively time consuming. For this reason, most field-scale simulations of miscible displacements are not carried out on high-resolution grids. Rather, one must have a proper scheme for scale-up of the high-resolution grid to coarser ones that can be used in the computations. A key issue with any upscaling procedure is how well the upscaled model replicates important aspects of the fine-scale flow behavior, for example, total injection and production rate, average pressure or saturation throughout the reservoir, and breakthrough times of injected flows. Additional issues are the degree of upscaling achievable by a given method, the level of robustness of the upscaled model (that is, its applicability to models with different global boundary conditions or well locations), and whether or not the method introduces modifications to the governing equations. We will come back to this problem in Chapter 15.

14

Immiscible Displacements and Multiphase Flows: Experimental Aspects and Continuum Modeling

Introduction

In this chapter, we turn our attention to immiscible displacements and multiphase flows in porous media and fractured rock. A large number of factors affect this class of phenomena, among which are capillary, viscous, and gravity forces, the viscosities of the two fluids and the interfacial tension separating them, the chemical and physical properties of the pores' or fractures' surface, that is, whether or not there are surface active agents, and whether the surface is rough, the morphologies of the pore space and the fracture network, and the wettability of the fluids. We begin our discussions by studying wettability, as it has a strong influence on the distribution of two or more immiscible fluids in a porous system.

14.1

Wettability and Contact Angles

The concept of wettability was already introduced in Chapter 4, where we presented a brief description of it and discussed its effect on the capillar pressure. We now expand that discussion and address the problem in detail. Generally speaking, the rock-fluid interactions are what is called wettability. It strongly affects flow of two or three immiscible fluids in a porous medium and their distribution in the pore space, conventional and enhanced oil recovery processes, and many other phenomena such as coating of various type of surface, and has been studied for a long time by petroleum engineers (Bartell and Miller, 1928; Owens and Archer, 1971; Salathiel, 1973; McCaffery and Bennion, 1974; Batycky *et al.*, 1981) and others. It is also known that the oil recovery process itself can alter reservoir wettability (Wagner and Leach, 1959; Reed and Healy, 1977). We will come back to the issue of wettability alteration later in this chapter.

Consider, as an example, an experiment in which a drop of water is placed on a surface immersed in oil. Then, a contact angle is formed that may vary anywhere from 0 to 180° (see Figure 4.6). As can be seen there, there are three distinct surface tensions that are related by the Young–Dure equation, Eq. (4.13), derived in

Chapter 4:

$$\sigma_{ow} \cos \theta = \sigma_{os} - \sigma_{ws} , \quad (14.1)$$

where σ_{ow} is the interfacial tension between oil and water, and σ_{os} and σ_{ws} are the surface tensions between oil and the surface, and water and the surface, respectively. Normally, the contact angle θ is measured through the water phase. Strictly speaking, if $\theta < 90^\circ$, the surface is considered as water-wet. In practice, however, $\theta < 65^\circ$ for water-wet surfaces, while $105^\circ < \theta < 180^\circ$ for oil-wet ones. If $65^\circ < \theta < 105^\circ$, the surface is said to be *intermediately-wet*. Such a surface has no strong preference for any of the two fluids. Another important concept is *mixed wettability*, in which the wettability of the surface changes from pore to pore (in a porous medium), or even from one portion of the surface to another. Mixed wettability is attributed to the *chemical heterogeneity* of the surface, and is actually the situation that one must deal with in most oil reservoirs.

The study of moving contact lines and contact angles goes back to Washburn (1921) who proposed Eq. (4.17) for a cylindrical tube. The Washburn equation is invalid if the length of the tube is much longer than its diameter, and if the diameter is small enough that gravity cannot have a significant effect on the shape of the moving contact line or meniscus. Fisher and Lark (1979) accumulated experimental evidence in support of the Washburn equation. However, Eq. (4.17) neglects the structure of the surface and its effect on a moving contact line; it only provides an overall picture of what happens.

In almost all cases of practical interest, one must address the problem of the no-slip boundary condition at the solid surface. Huh and Scriven (1971) were the first to analyze moving contact angles and lines and concluded that, with a no-slip boundary condition, the stresses at the contact line *diverge*. They attributed the divergence to the existence, among other things, of discontinuous processes around the contact line. As pointed out by Dussan V. and Davis (1974), however, the anomaly is due to the fact that Huh and Scriven (1971) had analyzed a *planar* interface, and that their equations failed to satisfy the normal stress boundary condition at the interface between the two fluids. It is now well-known that there are two ways of removing the singularity. If the advancing fluid wets the solid surface perfectly, then a thin precursor film forms *ahead* of the contact line and the dissipation divergence (which is logarithmic) has a cutoff at the film thickness. On the other hand, if the advancing fluid does not wet the surface completely, slip can occur within a length l_θ from the contact line, which acts as a cutoff and prevents the divergence of the total dissipation. These matters were discussed in detail by Dussan V. (1979).

On a moving contact line, the interface exerts a force $\sigma_{fs} \cos \theta_D$, where θ_D is the apparent *dynamic* contact angle (as opposed to a static contact angle θ_S , which is well-defined on a homogeneous surface), and σ_{fs} is the surface tension between the fluid and the solid. There is also an additional viscous force F_v on the contact line. If the capillary number, $Ca = \mu v / \sigma_{ow}$, is small, F_v would also be small compared with the capillary forces, except within a distance l_θ from the contact line. This fact provides us with a method of measuring F_v knowing θ_D , measured far from

the contact line since $F_v = \sigma_{fs}(\cos \theta_D - \cos \theta_S)$. Surface roughness, chemical heterogeneity and other factors make this picture more complex, which is discussed below. The dynamic contact angle θ_D can be measured by optical methods (Hoffman, 1975).

14.2

Core Preparation and Wettability Considerations

Experiments with cores (conventional or plugs) for determining capillary pressure curve, the relative permeabilities, and the wettability of porous samples remain the pillar of what is usually called *formation evaluation*. A review of core analysis techniques was given by Keelan (1972), which remains quite readable and relevant today. Before any core analysis, its objectives must be carefully defined because sometimes they may conflict and make it impossible to satisfy all the requirements on a given well in a field-scale porous medium, such as, an oil reservoir. Common objectives include securing cores for

1. estimation of the porosity, permeability, lithology, and residual fluid saturations (see below), and predictions of probable production, for example, of oil from the reservoir from which the core is obtained.
2. Identifying significant changes in the porosity, permeability, and lithology in order to characterize the porous medium.
3. Estimation of the interstitial water saturation (see below), and
4. samples of the porous medium in an unaltered wettability and/or saturation state for special tests.

Changes in the core may occur in its in-situ saturation during (1) the coring operations, and (2) the subsequent pressure reduction and gas expansion that occurs as the core is transferred from the reservoir temperature and pressure to the atmospheric conditions. The core's residual saturations of, for example, oil and water, determined in the laboratory reflect (1) the coring fluids used and, hence, the filtrate lost; (2) the degree of flushing; (3) the formation fluid properties; (4) the porous sample's relative permeability characteristics (see below); (5) core packing and preservation after removal from the core barrel to the time of analysis, and, (6) in some cases, the analytical techniques.

Filtrate loss during coring may displace some of the original fluids present in the rock. Coring fluids that are selected – water- and oil-based, inverted oil emulsion, hydrocarbon gas, and air – must reflect the objectives of the core analysis. Anderson (1986a,b, 1987a,b,c) and Hirasaki (1991) presented comprehensive discussions of the various parameters that affect the wettability of cores as well as the effect of wettability on various core properties.

The wettability of a porous medium affects almost all types of core analysis and the medium's properties, including the capillary pressure (see Chapter 4), relative permeability (see below), water flood behavior, irreducible water and residual oil

saturations (see below), the electrical properties, dispersion of tracers (see Chapters 11 and 12), and the results of any computer simulation for predicting tertiary recovery in the case of oil reservoirs. Thus, for core analysis to accurately predict the properties of a porous medium, the wettability of the core must be the same as that of the undisturbed porous medium.

Cores in three different states of preservation are used in core analysis, namely, native, clean, and, restored states. The term native state (sometimes called the fresh state) is used for any core that is obtained and stored by methods that preserve the wettability of the original porous medium and include cores that are obtained by both the oil-based and water-based drilling mud. To obtain a clean core, attempts are made to remove all the fluids and adsorbed organic material by flowing solvents through the core. Cleaned cores usually become strongly water-wet, and should only be used for such measurements as the porosity and single-phase permeability on which the wettability has no effect, and in tests when the porous medium is known to be strongly water-wet. In the restored cores, the native wettability is restored by (1) cleaning the core; (2) saturating it with brine followed by crude oil, and (3) aging the core at the porous medium's temperature for about 1000 h. Hirasaki *et al.* (1990) described the evaluation of the wettability using a restated core.

The wettability of originally water-wet mineral surfaces can be altered by the adsorption of polar components and/or the deposition of organic matter that was originally in the crude oil. Surfactants in crude oil are generally believed to be polar components that contain oxygen, nitrogen, and/or sulfur. Such components are prevalent in the heavier fractions of crude oil. Wettability alteration is determined by the interaction of the oil constituents, the mineral surface, and the brine chemistry including ionic composition and the pH. In silica/oil/brine systems, trace amounts of multivalent metal cations can alter the wettability. The cations reduce the solubility of crude oil surfactions and/or activate the adsorption of anionic surfactants onto the silica. The multivalent ions include Ca^{2+} , Mg^{2+} , Cu^{2+} , Ni^{2+} , and Fe^{3+} . Experiments indicate that the wettability of coal, graphite, sulfur, talc, talc-like silicates, and many sulfides ranges from neutrally wet to oil-wet. Most other minerals, including quartz, carbonates, and sulfates, are strongly water-wet in their native state. Unfortunately, however, many factors significantly alter the wettability of the core. Such factors are divided into two groups, namely, (1) those that influence core wettability *before* testing, such as drilling fluids, packaging, preservation, and cleaning, and (2) those that influence wettability *during* testing, such as test fluids, temperature and pressure.

The wettability of a core can also be altered during drilling by the flushing actions of drilling fluids, especially if the fluid contains surfactants or has a pH different from that of the porous medium's fluids. Three coring fluids are generally recommended for obtaining a native-state core that are (1) synthetic formation brine; (2) unoxidized lease crude oil, and (3) a water-based mud with a minimum additive. Because of surfactants in the system, no commercially available oil-based or oil-emulsion mud is known that preserve the native wettability. If the original wettability of a porous medium is not inadvertently modified, a native-state core would yield characteristics that are closest to those of the porous medium. Howev-

er, the necessary procedures to preserve the wettability are troublesome and time-consuming. Even after all the precautions are taken, it is still possible that the wettability is altered through oxidation or due to deposition of heavy components as the core is brought to the surface.

An important issue is the precise procedure that should be followed in order to obtain the most reliable information from cores in which the wettability is altered. As stated before, cleaned cores may be used in core analysis. Grist *et al.* (1975), Cuiec (1977), and Gant and Anderson (1988) studied core cleaning and restoration problems. One problem with a cleaned core is that it is sometimes difficult, if not impossible, to remove all the adsorbed materials. If this is the case, the wettability of the cleaned core may be in an unknown state, hence causing variations in core analysis. In many cases, a core is also contaminated by drilling mud surfactants. Moreover, any single solvent is relatively ineffective in core cleaning and better results are obtained with a mixture or a series of solvents. Specific combinations of chloroform, benzen, carbon disulfide, ethanol and toluene, together with crude oil and cores are known to yield a good result, though a poor result when used alone.

When only one core with altered wettability is available, the best possible multiphase measurements are obtained by restoring the porous medium's wettability with a three-step process, as described before. After cleaning and flowing reservoir fluids into the core sequentially, the core is aged at the formation temperature for a sufficiently long time to establish adsorption equilibrium. The aging can be at either the formation pressure with live crude, or at ambient pressure with dead crude oil. In the former case, the solubilities of the wettability-altering compounds should have their original values, whereas it is possible that the wettability differs when dead crudes at ambient pressure are used.

Once a native-, cleaned- or restored-state core is obtained, core analysis is performed. The tests are run with either crude or refined oil at ambient or formation temperature and pressure. Because the wettability effect is ignored, cleaned cores are generally analyzed with refined oil at room conditions. To preserve the wettability, the most accurate tests are run with native or restored cores at reservoir condition using live crude oil and brine because it is the best imitation of the conditions of a reservoir or porous medium. Note also that cores are generally more water-wet at the original conditions than they are at room temperature and pressure.

Anderson (1986a) described the effects of the experimental conditions on wettability, the most important of which are as follows.

1. *The formation temperature versus room temperature:* Changing the temperature tends to make the core more water-wet at higher temperatures due to (1) increasing the solubility of wettability-altering compounds, some of which even desorb from the surfaces as the temperature rises, and (2) decreasing the interfacial tension and the contact angle measured through the water. Such an effect has been seen in experiments with cleaned cores, mineral oil, and brine even though there were no compounds that could adsorb and desorb.
2. *Live versus dead crude at formation pressure:* The use of dead crude instead of live crude oil at ambient or reservoir pressure may also change the wettability be-

cause the properties of the crude are altered. Light ends are lost from the crude, while the heavy compounds are less soluble, which may make the core more oil-wet. In some experiments, pressure appears to be much less important than temperature.

3. *Refined versus crude oil:* Because refined oils are much easier to work with, it is a common laboratory practice to flush native- or restored-state cores with refined oil before carrying out any tests. There is, however, the possibility that doing so alters the wettability. It has been hypothesized that the desorption of wettability-influencing materials would require a correspondingly long period of time and, thus, the original wettability would be unaltered if laboratory tests using refined oil and brine were conducted quickly enough.

14.3

Measurement of Contact Angle and Wettability

Several methods of measuring the contact angles are in use. They include the tilting plate, the capillary rise, the tensiometric, the vertical rod, the cylinder, and the sessile drops or bubbles methods. Adamson and Gast (1997) described most of such methods. The petroleum industry, however, does not use most of them because they work best when one has pure fluids, and clean, artificial cores that are rarely encountered in practice. The methods that are used for such cores are described in detail by Anderson (1986a,b). Here, we restrict ourselves to describing three quantitative methods. More details are given by Anderson (1986b).

14.3.1

The Sessile Drop Method

The most popular method of measuring the contact angle in the petroleum industry is perhaps the sessile drop method (see, for example, McCaffery, 1972) and its modification by Treiber *et al.*, 1972. In the latter method, the mineral to be tested is put in a test cell that is made of inert material in order to prevent contamination of the surface that alters the true contact angle. Two flat and polished mineral crystals which are usually either quartz or calcite crystals (sedimentary rocks are composed of such crystals) are mounted parallel to one another. The apparatus must be completely clean so that the true contact angle is measured. The cell containing the mineral crystals is then filled with de-oxygenated synthetic formation brine. It usually takes a few days for the oil–crystal interface to be clearly established, a process that is called *aging*. Then, the two crystals are displaced parallel to each other to shift the oil drop. Thus, the brine can move over a portion of the surface that is covered with oil. In this way, an *advancing* contact angle θ_A is measured. Usually, it takes a day or two before θ_A reaches its equilibrium value. The surface is aged again, the water is advanced again, and so on. The sessile drop method uses the

same procedure, but with only one flat crystal. A drop of oil is formed at the end of a fine capillary tube and brought into contact with the flat surface (see Figure 4.6).

If the oil contains surface-active agents, θ_A increases with aging until adsorption equilibrium is reached that may, however, take a long time. Usually, the measured contact angles exhibit hysteresis, that is, the contact angle θ_A of an interface that was recently advanced differs from the apparent contact angle θ_R that recently receded. The hysteresis is presumably due to the existence of many metastable positions of the contact line. The difference $\theta_A - \theta_R$ can be as large as 60° . There are at least three reasons for the hysteresis that are (1) surface roughness; (2) surface heterogeneity, and (3) surface immobility on a macromolecular scale.

As pointed out by Morrow (1970, 1976), surface roughness and pore geometry affect the contact line between the two fluids and the surface and, thus, change the apparent contact angle. If the surface is smooth, then θ is fixed. In many natural porous media such as oil reservoirs, however, there are sharp edges that give rise to a wide range of contact angles. Figure 14.1 demonstrates the phenomenon clearly. The contact angles θ_A and θ_R were measured on a roughened teflon surface, while the intrinsic angle θ_E was measured on a smooth teflon surface. The contact angles were changed by varying the salinity. We shall return to this phenomenon shortly. Moreover, the compositional heterogeneity of a surface gives rise to θ_A and θ_R that can change from pore to pore, and contact angle measurements cannot take such effects into account.

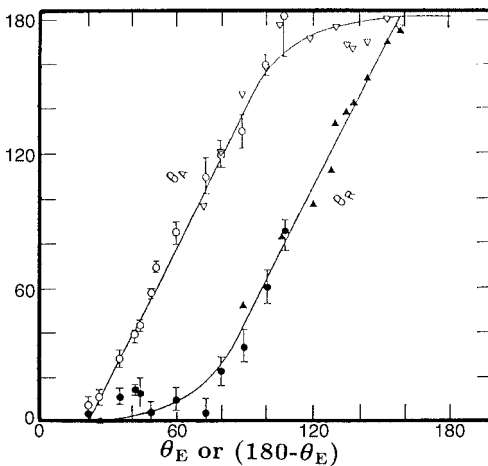


Figure 14.1 Recently advanced and recently receded contact angles (in degrees) θ_A and θ_R measured on roughened teflon, versus intrinsic contact angle θ_E measured on smooth

teflon. Open circles (triangles) represent advancing θ_E ($180^\circ - \theta_E$), while solid circles (triangles) are for receding angles (after Morrow, 1976).

14.3.2

The Amott Method

In the method developed by Amott (1959), a core is prepared by centrifuging with brine until the residual oil saturation (ROS) is reached, which is that volume fraction of oil in the pore space, residing in isolated finite clusters of pores, which can no longer be displaced by brine in a centrifuge. Then, four steps are taken for measuring the wettability of the core. (1) The core is immersed in oil, and the volume of water displaced by free or spontaneous imbibition of oil (see below) into the core is measured in a period of time that may be as long as one or two weeks. (2) The core in oil is centrifuged until the irreducible water saturation (IWS) – water saturation that can no longer be reduced by centrifuging – is reached and the total amount of displaced water is measured. (3) The core is immersed in brine, and the volume of oil displaced by water is measured. (4) The core is centrifuged in brine until the ROS is reached, and the total amount of displaced oil is measured.

To determine whether the core is water- or oil-wet, two quantities are calculated. One is $R_o = V_{wi} / V_{wt}$, where V_{wi} is the water volume displaced by free imbibition of oil, and V_{wt} is the total volume of water displaced by free and forced (centrifugal) displacements. The second quantity is $R_w = V_{oi} / V_{ot}$, where the notations have similar meanings. Now, if the porous medium is water-wet, then $R_w > 0$ and $R_o = 0$ because in a water-wet porous medium, there can be no free imbibition of oil (the oil must be forced into the medium), and vice versa. In a sense, the method measures the average wettability of the sample. Sometimes, an index, $I_w = R_w - R_o$, is measured that can vary anywhere from +1 (completely water-wet porous media) to -1 (completely oil-wet porous media). According to Cuiec (1984), for $0.3 \leq I_w \leq 1$, the medium must be considered as water-wet, for $-0.3 \leq I_w \leq 0.3$ it is intermediately-wet, whereas for $-1 \leq I_w \leq -0.3$, it is oil-wet. The main problem is that if a porous medium is close to being intermediately-wet, then the method is not very sensitive or accurate simply because free imbibition of either fluid cannot take place in significant amounts.

14.3.3

US Bureau of Mines Method

This method was developed by Donaldson *et al.* (1969). Similar to the Amott method, it measures the average wettability of the system and is based on comparing the work necessary for one fluid to displace the other. The wetting fluid requires less work to displace the nonwetting fluid from the core than the opposite. It can be shown (Morrow, 1970) that the required work is proportional to the area under the capillary pressure curve in a capillary pressure–water saturation plot (see Chapter 4). If the porous medium is strongly water-wet, most of the water will imbibe freely into the core, and the area under the curve for water will be very small. Thus, the capillary pressure curves for the two displacements are measured, and a wettability index, $I_w = \log(A_o/A_w)$, is calculated where A_o and A_w are the areas under the oil- and water-drive curves, respectively. Clearly, if $I_w > 0$, then the

porous medium is water-wet, and if $I_W < 0$, it is oil-wet. If $I_W \simeq 0$, then the porous medium is close to being intermediately-wet. For most natural porous media, such as oil reservoirs, $-1.5 \leq I_W \leq 1$.

14.4

The Effect of Surface Roughness on Contact Angle

We already briefly described the experimental work of Morrow (1970, 1976), who studied the effect of surface roughness on contact angles. There have also been a few theoretical studies of the effect of a heterogeneous surface on moving contact angles and contact lines (Joanny and de Gennes, 1984; Pomeau and Vannimenus, 1985; Joanny and Robbins, 1990). Joanny and Robbins (1990), for example, studied the motion of a contact line on a surface with periodic heterogeneities. Although such heterogeneities do not usually exist on natural surfaces, their study provides clues as to how the phenomenon may be studied on a real heterogeneous surface. They considered the case in which the contact line is advanced by a constant force F or a constant velocity v . In the first case, the motion starts if $F > F_{C1}$, where F_{C1} is some threshold force that is related to the static contact angle θ_A or θ_R defined earlier. For smooth heterogeneities, they found that $F - F_{C1} \sim v^2$. If the contact line is moved at a constant velocity, the results are somewhat different: one has two regimes, namely, the weak and strong *pinning* regimes in which the interface is pinned to the solid surface and does not move. In the strong pinning regime, which is more interesting and relevant, there is another threshold F_{C2} that approaches F_{C1} as the surface becomes more heterogeneous. For smooth heterogeneities, they obtained $F - F_{C2} \sim v^{2/3}$. Similar results were obtained by Raphael and de Gennes (1989).

14.5

Dependence of Dynamic Contact Angle and Capillary Pressure on Capillary Number

Stokes *et al.* (1990) studied the velocity dependence of $\cos \theta_D - \cos \theta_S$ by a method that, in a sense, represents one for contact angle measurements, except that it is more sensitive and its details are also different from those of the methods described earlier. They measured the capillary pressure P_c across the interface as a function of the contact-line velocity v by superimposing a small-amplitude oscillatory flow on a larger steady-state flow and measuring the response. The steady-state flow was varied by raising a reservoir of the advancing fluid. Between the reservoir and the sample, a narrow-bore and long tube were inserted to guarantee that the flow rate was constant. The interface between a mineral oil and a glycerol-methanol mixture was measured in a 1-mm diameter, 30-cm long Pyrex tube. Two Omega pressure transducers were used to measure both the pressure and velocity. The AC (alternating current) output of the transducers was amplified, and the harmonic content measured with several amplifiers. When the glycerol mixture was advanced,

$\theta_A = 65^\circ$ and $\theta_R = 45^\circ$ were measured, which indicated that the surface of the tube was somehow disordered. The data indicated that for a tube of radius R ,

$$\cos \theta_D - \cos \theta_A = \frac{R}{2\sigma_{fs}} (P_c - P) = aCa^x, \quad (14.2)$$

where P is the total pressure, $a \simeq 3.1 \pm 1$, and $x \simeq 0.4 \pm 0.05$. Equation (14.2) implies that $P_c \sim v^x$. For *homogeneous* surfaces, the velocity dependence of θ_D has been studied extensively. For example, Cox (1985) found that for θ_D measured at a distance r from the contact line, one has

$$g_\theta(\theta_D, M) = g_\theta(\theta_0, M) + \left[\ln \left(\frac{r}{\ell_\theta} \right) + B_1 \right] Ca, \quad (14.3)$$

where g_θ is a simple analytic function, M is the viscosity ratio of the two fluids, θ_0 is the actual contact angle at lengths smaller than ℓ_θ , and B_1 is a constant that depends on the model. Molecular dynamics simulations of Koplik *et al.* (1988a) confirmed the validity of Eq. (14.3) if one takes $\theta_0 = \theta_S$. Equation (14.3) implies $x = 1$ for an intermediately-wet fluid on a homogeneous surface, which is different from what Eq. (14.2) implies. The difference may be attributed to the roughness or heterogeneity of the surface, an effect that is apparently strong enough to change the exponent x from unity to about 0.4. The difference also indicates the significance of surface roughness and its effect on the contact angle (see also below).

On the other hand, Rillaerts and Joos (1980) were able to correlate data for several different systems by plotting $\cos \theta_D - \cos \theta_S$ versus $Ca^{1/2}$, which is close to Eq. (14.2). Hoffman (1975) proposed somewhat more complex correlations that can describe many different sets of data for all $0^\circ \leq \theta_D \leq 180^\circ$. For example, at low values of Ca , he obtained $\theta_D \sim Ca^{1/3}$, which is known as the *Tanner's law*. Weitz *et al.* (1987) also measured the velocity dependence of the capillary pressure (and, hence, θ_D) between two fluids in a *porous medium* and proposed the correlation

$$P_c \simeq \frac{\sigma_{fs}}{r_t} (B_2 Ca^x - 1), \quad (14.4)$$

where r_t is a typical radius of pore throats of the porous medium. Their measurements yielded $x \simeq 0.5 \pm 0.1$ and $B_2 \simeq 300$, which are compatible with the results of Stokes *et al.* (1990). de Gennes (1988) used scaling analysis to propose that

$$P_c \simeq \frac{\sigma_{fs}}{r_t} Ca^{\frac{2}{3}} \tau^{\frac{5}{3}}, \quad (14.5)$$

where τ is a tortuosity factor. Although Eq. (14.5) is almost consistent with Eq. (14.4), it does not seem to agree with the result of Stokes *et al.* There is, however, one major difference between the experiments of Stokes *et al.* (1990) and Weitz *et al.* (1987): the former were done in a *tube*, whereas the latter were performed in a *porous medium*.

14.6

Fluids on Rough Self-Affine Surfaces: Hypodiffusion and Hyperdiffusion

In Chapters 4 and 6, we described the self-affine fractal properties of pore and fracture surface. Fractality of pore surfaces has interesting implications for fluid distributions at low wetting-phase saturations. The problem was studied by de Gennes (1985), Melrose (1988), Davis (1989), Davis *et al.* (1990), and Toledo *et al.* (1990, 1992). We assume that one of the fluids strongly wets the surface so that even at very low saturations, the wetting phase remains hydraulically connected through thin films that are formed on the surface. The capillary pressure is given by the Young–Laplace equation

$$P_c = 2\mathcal{H}\sigma_{ff} , \quad (14.6)$$

where $\mathcal{H}$ is the mean curvature of the interface between the two phases, and σ_{ff} is the interfacial tension between the two fluids. Equation (14.6) is valid when both phases are present in large amounts. If, however, the wetting phase is present only in the form of thin films, then one must use the *augmented* Young–Laplace equation according to which

$$P_c = 2\mathcal{H}\sigma_{ff} + \Pi(h) , \quad (14.7)$$

where h is the film's thickness, and $\Pi(h)$ is called the *disjoining pressure*. At saturations below the percolation threshold, the wetting phase exists as thin films or *pendular structures* at intergranular contacts, or in nooks and crannies provided by the pore surface features or overhangs. Thus, at a given capillary pressure, the liquid volume is proportional to r^{3-D_f} , and because $r \sim 1/P_c$, we must have

$$S_w \sim P_c^{D_f-3} , \quad (14.8)$$

where D_f is the fractal dimension of the pore space (see Chapter 4).

Davis (1989) analyzed Melrose (1988)'s data and found that they are well-described by Eq. (14.8) with $D_f \simeq 2.55$ (see Figure 14.2). His estimate of D_f is in the range of fractal dimensions reported by Katz, Thompson, Krohn and others that were described in Chapter 4. If the capillary contribution, $2\mathcal{H}\sigma_{ff}$, is small compared with the disjoining pressure, then $P_c \simeq \Pi(h) \sim h^{-m}$ and, therefore,

$$S_w \sim P_c^{-\frac{1}{m}} . \quad (14.9)$$

Similar relations may be developed for the hydraulic conductivity g_h of the wetting phase. Thus, if only thin films are present in the pores, then, since $g_h \sim h^3$, we find that

$$g_h \sim P_c^{-\frac{3}{m}} \sim S_w^3 . \quad (14.10)$$

However, if only the pendular structures are present and distributed fractally, then

$$g_h \sim S_w^{\frac{3}{m(3-D_f)}} . \quad (14.11)$$

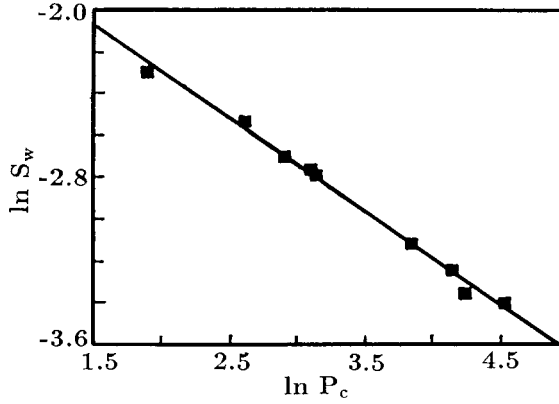


Figure 14.2 Fractal analysis of Melrose's data for the wetting-phase saturation S_w versus the capillary pressure P_c (after Davis, 1989).

If a porous medium with a low wetting-phase saturation is immersed in a reservoir of a wetting fluid, it will spontaneously imbibe the wetting phase. The saturation of the wetting phase will satisfy a convective-diffusion (CD) equation in which the dispersion coefficient D_L is given by

$$D_L^{(c)} = -\frac{g_h}{\mu_w} \frac{dP_c}{dS_w}, \quad (14.12)$$

where μ_w is the viscosity of the wetting phase. $D_L^{(c)}$ is usually called the *capillary dispersion coefficient*, and is not the same as the longitudinal dispersion coefficient D_L introduced and studied in Chapters 11 and 12. Depending on the wetting-phase saturation, one can have three distinct regimes.

1. If $D_L^{(c)} \rightarrow 0$ as $S_w \rightarrow 0$, the invading front of the wetting phase will disperse less than that in a diffusive front. This is called *hypodiffusion*.
2. If $D_L^{(c)}$ approaches a constant as $S_w \rightarrow 0$, then we have a diffusive dispersal of the front.
3. If $D_L^{(c)} \rightarrow \infty$ as $S_w \rightarrow 0$, then the front will disperse faster than that in a diffusive propagation and is called *hyperdiffusion*. From Eqs. (14.9)–(14.12), we obtain

$$D_L^{(c)} \sim S_w^\alpha, \quad \alpha = \frac{3 - m(4 - D_f)}{m(3 - D_f)}. \quad (14.13)$$

Therefore, if $m < 3/(4 - D_f)$, we will have capillary hypodiffusion; if $m > 3/(4 - D_f)$, the regime of capillary hyperdiffusion is operative, whereas $m = 3/(4 - D_f)$ gives rise to a capillary diffusive front.

Bacri *et al.* (1985) carried out experiments in which water imbibed in a pre-wet sandstone (i.e., a porous medium at its IWS), and reported hyperdiffusion. Bacri

et al. (1990b) studied the same problem in a porous medium made of glass beads which are totally wetted by water, and in another porous medium consisting of polymethylmethacrylate (PMMA) beads that are partially wetted by oil and water. In the porous medium with glass beads, they observed hyperdiffusion again, whereas PMMA beads did not allow hyperdiffusion to happen. They also studied the same phenomenon in a porous medium with a mixture of the two types of beads and reported hyperdiffusion, even when the fraction of glass beads was small (but not smaller than the critical volume fraction for percolation).

Using Eqs. (14.8)–(14.10), Toledo *et al.* (1990) analyzed the data of Nimmo and Akstin (1988) who had reported measurements of P_c and g_h at low saturations in the presence of air in several compacted samples of Oakley sands, a clayey soil. On the other hand, Viani *et al.* (1983) provided data on the disjoining pressure of clayey soils, which indicated that they were well-described by $\Pi(h) \sim h^{-1/2}$, implying that $m = 1/2$. Toledo *et al.* (1990) showed that the power laws hold for nine different samples with a fractal dimension $2.35 < D_f < 2.67$.

Davis *et al.* (1990) also analyzed Ward and Morrow (1987)'s data on capillary pressure–saturation curves in the presence of air in several low-permeability sandstones. Two distinct regimes were identified. One was in the lowest saturation region for which $D_f \simeq 2$. The second one was in a higher saturation region for which $2.6 \leq D_f \leq 2.9$, which is consistent with the range of fractal dimensions discussed in Chapter 4. Finally, Novy *et al.* (1989) developed a pore network model of two-phase flow in porous media to test the above power laws, and reported qualitative agreement between their simulations and the power laws.

The wettability of a porous medium also has a strong effect on its transport and thermodynamic properties. There is a strong correlation between the shape of the capillary pressure curves of a medium and its transport properties. Thus, we first describe the effect of wettability on capillary pressure curves.

14.7

Effect of Wettability on Capillary Pressure

Let us first introduce the terminology that is frequently used in this and the next chapter. In *drainage*, a non-wetting fluid displaces a wetting one from a porous medium, where *imbibition* is a process by which a wetting fluid displaces a non-wetting fluid. Using sintered porous teflon and clean fluid pairs, Morrow (1970, 1976), McCaffery and Bennion (1974), and Morrow and McCaffery (1978) studied two-phase relative permeabilities (see below) and capillary pressure curves in porous media that are uniformly wettable. They took, as an independent measure of the wettability, the intrinsic contact angle θ_E made by the fluid pairs on smooth teflon surfaces and studied two processes. One was *primary displacement* – the reduction of the saturation of a reference fluid from 100% to the residual saturation (RS) by injection of a non-reference fluid into the porous medium. The second process was *secondary displacement* that follows the primary one, that is, reduction of the non-reference fluid saturation to the RS by injection of the reference flu-

id. On the basis of the shape of the capillary pressure (and relative permeability) curves in the sequence of primary and secondary displacements, they identified three regimes of wettability:

1. *wetted*, in which the primary displacement is drainage and secondary displacement is imbibition;
2. *intermediately-wetted*, in which the primary and secondary displacements are both drainage, and
3. *non-wetted*, in which the primary displacement is imbibition, whereas the secondary displacement is drainage.

The second case is very interesting. Since the primary displacement is drainage, one intuitively expects the secondary displacement to be imbibition. In the intermediate wettability regime, however, the operative contact angle appears to give the more strongly wetting characteristics to the fluid being displaced, regardless of whether it is the reference fluid or the non-reference one.

Figure 14.3b presents the capillary pressure curves for a typical wetted regime measured by Killins *et al.* (1953). The measurements were done using a water-wet Berea sandstone. The primary process, denoted by one on the curve, is drainage in which oil displaced water and was terminated at A, where the IWS was reached. This was followed by process two, a spontaneous imbibition of water into the core up to point B at which $P_c = 0$. Beyond B, the water must be forced into the medium (curve 3), characterized by a negative P_c , until point C is reached at which the ROS is reached. Note the two typical knees, one at the beginning of drainage, and the second one at B.

Figure 14.3b should be compared with Figure 14.3a that is typical of capillary pressure curves in non-wetted porous media. The data were measured in an oil-wet Berea sandstone that had been treated with Drifilm in order to render it strongly oil-wet. Note that in both processes, one – spontaneous imbibition of oil – and two – drainage of oil by water – the capillary pressure curve takes on negative values. Moreover, the imbibition curve rises steeply, whereas the drainage curve proceeds slowly, except near the original starting point A. Finally, Figure 14.3c shows schematic capillary pressure curves for typical intermediately-wet systems.

In order to compare capillary pressure data measured with different cores, though from the same porous medium, and to take into account the effect of the permeability and porosity of each core, the curves are usually replotted in terms of the Leverett J -function, Eq. (4.29). As long as the wettability of all cores is the same, the $\cos \theta$ term of Eq. (4.29) is a simple numerical factor. However, if different fluids are used with cores from the same porous medium or reservoir, then the term becomes important. Figure 14.4 presents the effect of contact angle on capillary pressure, reported by Morrow (1970). As the contact angle increases, the return curve of the capillary pressure (to the left of the dashed curve) becomes steeper, which is in agreement with Figure 14.3c because increasing θ implies the tendency of the system towards intermediate wettability. In fact, in the last curve

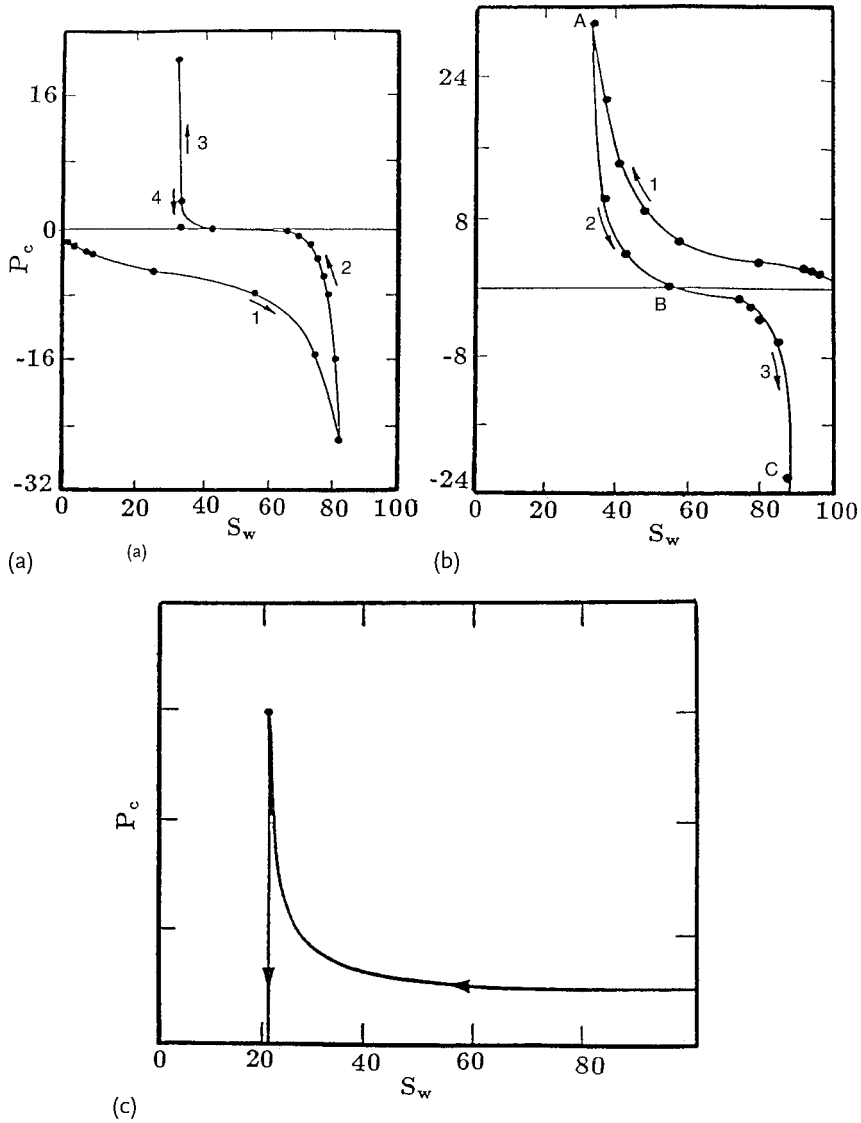


Figure 14.3 (a) Typical capillary pressure P_c (in cmHg) versus saturation S_w (in percent) for a non-wetted regime; (b) for a wetted porous medium (after Killins *et al.*, 1953), and (c) for an intermediately-wet system. Curve 1 represents drainage, 2 represents spontaneous imbibition, 3 denotes forced imbibition, and curve 4 is the secondary drainage.

on the left (shown by ■), the advancing contact angle θ_A is about 77° that, as pointed out above, is well within the range of intermediate wettability.

There is yet another wettability regime that is of great practical importance. This is the so-called *mixed wettability* regime (Owens and Archer, 1971; Treiber *et al.*,

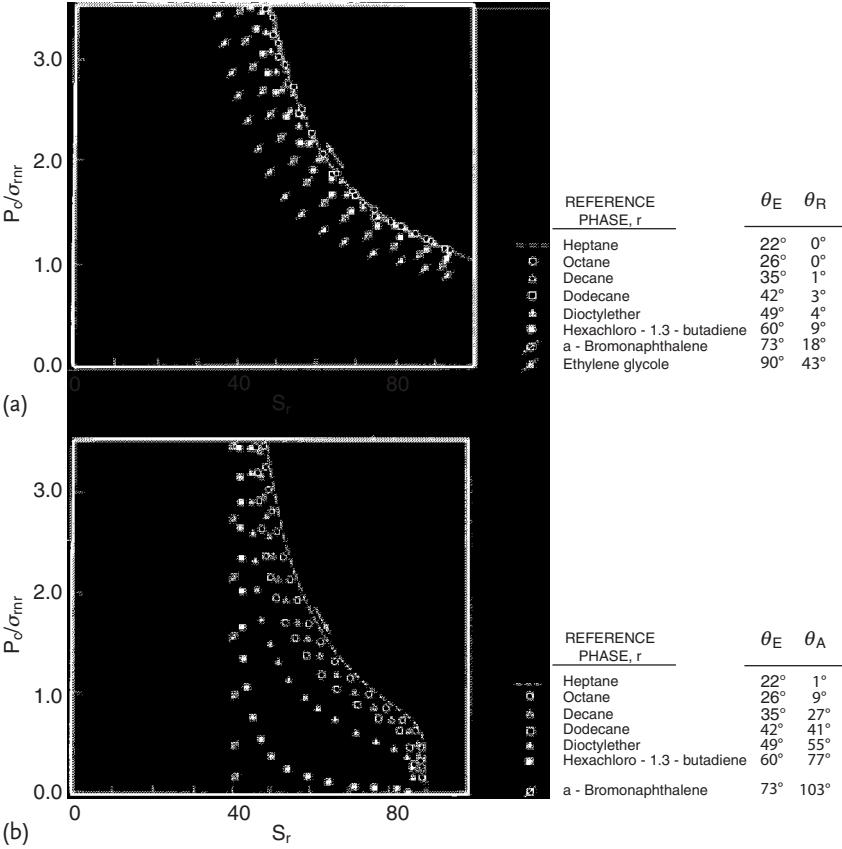


Figure 14.4 (a) Effect of contact angle on capillary pressure P_c divided by the surface tension P_c/σ_{nr} as a function of the saturation S_r of the reference fluid (in percent). P_c/σ_{nr} is in cm^{-1} . (a) Injection of the fluids. (b) Retraction of the fluids (after Morrow, 1970).

1972; Salathiel, 1973; Craig, 1971) in which some pores are wetted by one fluid, while others are wetted by the other fluid. There is evidence that on the scale of geological times, oil displaces brine from a portion of the pore space. Depending on the nature and composition of the oil, those portions of a pore wall that are separated from the crude oil by only a thin film or a tiny pendular structure of brine, may become oil-wet owing to diffusion and adsorption or stronger chemical interactions with constituents of the oil. One widely cited interaction is the deposition by the crude of polar organic surfactants upon the surface of the rock (Benner and Bartell, 1941; Salathiel, 1973; Melrose, 1988). It appears that migration, accumulation and deposition processes can generate a distribution of surface wetting preferences with the small pores that have not been invaded by oil remaining water-wet, whereas the larger pores that have been occupied by oil becoming more or less oil-wet. Figure 14.5 presents qualitative patterns of the capillary pressure curves for

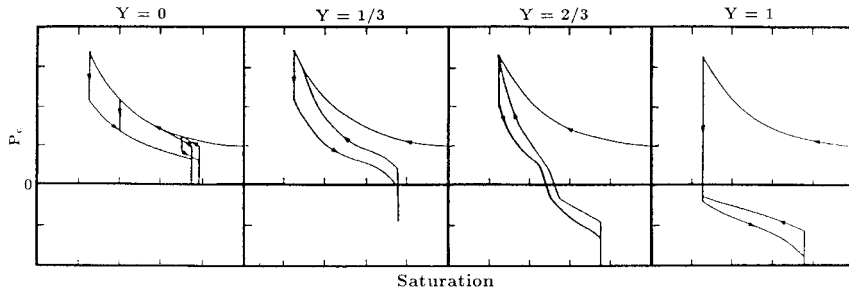


Figure 14.5 Capillary pressure curves for porous media with mixed wettability. Y is the fraction of the oil-wet pores (after Heiba, 1985).

a porous medium in which a fraction Y of the pores was invaded by oil and became oil-wet. $Y = 0$ corresponds to a totally water-wet system, while $Y = 1$ is representative of a totally oil-wet porous medium. More extensive discussions of the effect of wettability on capillary pressure curves were given by Melrose (1965, 1968) and Anderson (1987a).

14.8

Immiscible Displacement Processes

Having explained the most important factors that influence multiphase flow in porous media, we are now ready to describe the displacement of one fluid by another immiscible fluid in a porous medium. The displacement process is controlled and affected by a variety of factors that were mentioned at the beginning of this chapter. We already described the effect of the wettability and contact angles on capillary pressure, and will describe their effect on two-phase flow and transport properties of a porous medium later in this chapter. Among the remaining factors, the capillary number Ca and the mobility ratio M are of the greatest importance. Depending on how the displacement process proceeds, many regimes may arise. A careful discussion and classification of imbibition processes and how to distinguish between them was given by Payatakes and Dias (1984). Here, we present a summary of their classification and discussion, expanding on them if appropriate.

1. *Spontaneous imbibition*, which we already mentioned in the context of capillary pressure curves studied in Chapter 4.
2. *Constant influx, constant capillary number imbibition*, which occurs if a pressure drop ΔP is applied to a porous medium and is adjusted as the invading fluid expels more of the in-place fluid from the medium. If the mobility ratio $M \leq 1$, then we must maintain $\Delta P \geq 0$, and vice versa. If for $M \leq 1$, the applied pressure dominates the capillary forces, we will no longer be dealing with an imbibition process.

3. *Quasi-static imbibition*, which happens when the flow rate of the displacing fluid is vanishingly small. In this case, the interface between the displacing and displaced fluid advances only *one* pore at a time.
4. *Dynamic invasion with constant flow rate of the displacing fluid*, which can be carried out at either a favorable mobility ratio ($M \leq 1$), or at an unfavorable one ($M > 1$). To achieve the displacement, a large pressure drop ΔP is applied to the porous medium, which can be so large that it would dominate the capillary forces. The interface advances in *many* pores at any given time. For $M > 1$, viscous fingers can develop (see Chapter 13).

Note that the reverse of at least some of the imbibition processes may be considered as drainage processes. At the end of the displacement processes, the displaced fluid only exists in isolated blobs or clusters of finite sizes that can no longer be displaced by any of the above displacement processes. In order to mobilize and displace such blobs, the capillary number must be significantly increased, which would then give rise to three other displacement processes that are (Payatakes and Dias, 1984) *quasi-static* and *dynamic displacement* of blobs, both of which are time-dependent phenomena, and *steady-state dynamic displacement*. The last process can be carried out if the displacing and displaced fluids are *simultaneously* injected into the porous medium. After some time, a dynamic equilibrium is reached and the macroscopic flow rate becomes constant. This problem was studied by Hashemi *et al.* (1998, 1999a,b) using computer simulations, and was discussed from an experimental view point by Craig (1971). Payatakes (1982) presented excellent discussions of blob mobilization and displacement in porous media. Therefore, we do not describe and discuss the last three displacement processes in detail.

14.8.1

Spontaneous Imbibition

The driving force for spontaneous imbibition is capillary suction because of which the smallest pore bodies that are next to the interface are always invaded first. At any given time, many pore-level interfaces advance in the porous medium. Usually, the displacement takes place at small but finite capillary numbers Ca . Moreover, the value of Ca , which depends on the extent of the process, does not remain a constant, but varies over a range of values. This was nicely demonstrated in the experiments of Legait and Jacquin (1982) who studied spontaneous imbibition of water in sandstones containing oil at $M = 225$. They reported that the rate of oil displacement increased strongly with time. When the same experiment was carried out at $M = 1$, the same phenomenon occurred, albeit in a much weaker manner.

When relatively large values of Ca are created, a transition zone develops in which there is a high saturation gradient. As the interface advances in the medium, two separate regions develop. One is in front of the transition zone in which the saturation of the displaced fluid is high. The second one moves behind the transition zone in which the saturation of the displacing fluid is high and continues to expand as the interface advances. The transition zone remains essentially the

same throughout the displacement, except when the interface nears the end of the porous medium. For this reason, this region is called the *stabilized zone*. Although Bail (1956) had argued that under conditions used in regular imbibition in oil reservoirs, the length of this zone is not very large, its dynamics are interesting because they affect the efficiency of the displacement. It is also in this region that oil blobs are formed by disconnection of the displaced fluid by the displacing fluid.

We should point out that one of the main assumptions in models of multiphase flow through porous media, and in particular spontaneous imbibition, is that local capillary equilibrium is reached instantaneously. The assumption has been tested by both experiments and numerical simulations, but the results have been in conflict. For example, Li *et al.* (2003) contend that the assumption is valid, whereas Silin and Patzek (2004) and Le Guen and Kovscek (2006) contend the opposite. Note that if the nonequilibrium effect is truly present, then, at least over certain time scales, the RPs and many other important quantities will be time-dependent, and one would have to rederive new governing equations for multiphase flow in porous media (see below).

14.8.2

Quasi-Static Imbibition

One main difference between quasi-static and spontaneous imbibition is that at any given step of the former, only one pore is invaded by the displacing fluid, which is achieved by adjusting the backpressure so that the *narrowest* pore throat is invaded, while the interface at other larger throats remains essentially motionless. Since even the largest pore throats are smaller than the pore body to which they are connected, once a pore body is invaded, all the throats that are connected to it are also invaded. As soon as the interface enters such throats, the smallest pore body that is connected to them is invaded, and so on. Thus, at any given step of the displacement, the smallest pore body that is accessible from the external surface through a continuous path of the displacing fluid is invaded.

When the displacing fluid forms a sample-spanning cluster of invaded pore bodies (and the associated pore throats), the *breakthrough* occurs. Just before the breakthrough the displaced fluid is mostly connected. As the displacement proceeds, however, small blobs of the displaced fluid are formed that become trapped. At the end of the process, one may end up with a large number of isolated blobs with a significant volume fraction, the value of which depends on the morphology of the pore space. In unconsolidated porous media it varies between 0.14 and 0.2 (Chatzis *et al.*, 1983), whereas in consolidated media, it is anywhere between 0.4 and 0.8 (Wardlaw and Cassan, 1978).

Experimental data of Raimondi and Torcaso (1964), Egbogah and Dawe (1980), and Chatzis *et al.* (1983) indicate that the size distribution of the blobs, when expressed in terms of the number n_b of pore bodies that they occupy (recall that most of the volume of a pore space is provided by its pore bodies), satisfies the following

power law

$$n_b(s) \sim s^{-\tau}, \quad (14.14)$$

where $n_b(s)$ is the number of blobs of size s . Equation (14.14) is reminiscent of a similar the power law for the number of finite percolation clusters of s sites (see Chapter 3). Indeed, as we describe below, an appropriate percolation model can be devised to model such a quasi-static displacement. Egbogah and Dawe (1980) found that the size of the blobs varied between one and ten grain volumes, but most of them were around $s = 1$.

14.8.3

Imbibition at Constant Flow Rates

This process is very similar to spontaneous imbibition, except that in this case, we need to adjust a backpressure in order to keep the flow rate constant. There has been some controversy regarding the role of M in the displacement processes. Some early papers (for example, Geffen *et al.*, 1951; Donaldson *et al.*, 1966) claimed that for fluids with identical wettability characteristics, M does not have any significant effect. However, Le Febvre du Prey (1973) systematically investigated the effect of M on relative permeabilities to two-phase flows by studying displacements in sintered porous media in which the Ca varied between 10^{-7} and 5×10^{-3} . He found that the higher the viscosity of one of the fluids, the lower the relative permeability of the second fluid, with the effect being even more important than the wettability effect. The phenomenon is presumably due to the fact that the fluid with a high viscosity gives rise to a film that resides on the pore surface, which denies pore volume to the second fluid and decreases its relative permeability. Abrams (1975) found that the ROS in short porous media correlates well with the group $M^{-0.4}Ca$, where $10^{-7} \leq Ca \leq 10^{-2}$. Egbogah and Dawe (1980) found that for $M \leq 1$, the blob size distribution became much broader, took on bimodal and even trimodal shapes, and the average blob size increased dramatically.

14.8.4

Dynamic Invasion at Constant Flow Rates

The driving force for dynamic invasion is an applied pressure drop since the role of the capillary forces is of secondary importance. If $M > 1$, we will have an unstable displacement that will be described later. For now, we only consider the $M < 1$ case. There is a small transition zone in this process in which the saturation of both phases changes with time. If the capillary pressure is negligible compared with the applied pressure, we will have several advancing interfaces in as many pores at any given stage of the displacement. Because the driving force is the applied pressure, the microscopic interfaces choose the *largest* accessible pore throats (to minimize the resistance). Thus, the structure of the sample-spanning cluster of the displacing fluid resembles that in a drainage process. This does not necessarily mean that the smaller throats will not be selected: local pressures are also important and can

cause their invasion by the advancing fluid. Since in the transition zone the saturation of the fluids changes with time, the value of Ca cannot remain constant, even though the flow rate can be kept constant.

As in the previous cases, the advancing fluid creates isolated blobs of the displaced fluid. Whether the blobs become stranded or not depends on many factors. Ng and Payatakes (1980) and Payatakes *et al.* (1980) argued that the stranding of the blobs depends on Ca , the length of the blob in the direction of the macroscopic flow, and the sizes of the pore bodies and pore throats in which the blobs reside. If a very large blob is created, initially it is mobile, but later it breaks into several smaller blobs, with the breaking process continuing until the blobs are small enough to be stranded.

14.8.5

Trapping of Blobs

If the displaced fluid is incompressible, then at the end of both imbibition and dynamic invasion, one obtains many isolated blobs or clusters of the displaced fluid, the displacement of which is the main goal of oil recovery processes. The most common way of correlating experimental data for the trapped oil is in terms of a relationship between the residual wetting phase saturation S_{rw} , or that of the non-wetting phase saturation S_{rnw} , and a local capillary number Ca_l . Such a relationship is usually called the capillary desaturation curve (CDC). If Ca_l is small, then both S_{rw} and S_{rnw} are roughly constant. As Ca_l increases, one reaches a typical knee in the curve, beyond which the residual saturations decrease. Most waterflooding processes – those in which water is injected into an oil reservoir to displace the remaining oil – are well onto the plateau of a CDC where, as a general rule, $S_{rw} < S_{rnw}$.

In most cases, the two CDCs are normalized by their corresponding plateau values. Figure 14.6 shows a CDC constructed on the basis of experimental data obtained by several groups. Observe that the CDC for the wetting phase is to the right of that for the non-wetting fluid, which is not only typical, but also indicative of the significance of the wetting properties of porous media. Moreover, for some value of Ca_l , the two curves intersect, corresponding to some intermediate wetting condition. It also has been observed that the pore size distribution of the pore space can have a strong influence on the CDC. For a pore space with a broad pore size distribution, for example, a carbonate rock (see Chapter 4), the horizontal part of the CDC is very small or non-existent, whereas for a typical sandstone, one observes a significant horizontal part such as that shown in Figure 14.6.

Another important problem in which blob entrapment is important is CO_2 storage in brine aquifers (Ide *et al.*, 2007). Supercritical CO_2 is injected into a brine aquifer, and the resulting buoyant CO_2 plume migrates vertically upwards. The leading edge of the plume displaces the brine (as in drainage), whereas in its trailing region, brine displaces CO_2 (as in imbibition) which leads to the formation of entrapped CO_2 blobs or ganglia.

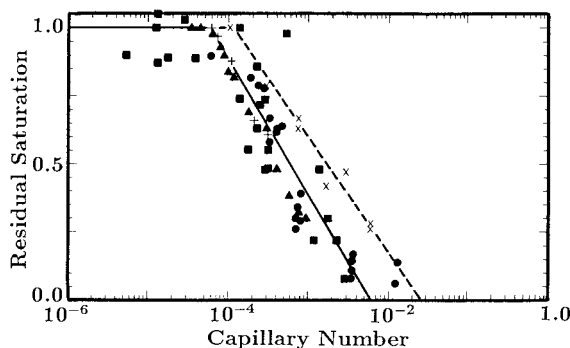


Figure 14.6 Capillary desaturation curves compiled with the available data for wetting fluids (dashed lines) and non-wetting fluids (solid line). The porous medium is a Berea core.

14.9

Mobilization of Blobs: Choke-Off and Pinch-Off

Mobilization of trapped oil blobs has been a problem of long-standing interest. Since the blobs have a size distribution, the residual saturation is only a fraction of the actual non-wetting saturation. Avraam *et al.* (1994) and Avraam and Payatakes (1995) carried out experiments over a wide range of parameters for quasi-steady-state flow of oil and water and showed that the entrapped oil blobs significantly contribute to the flow of oil. They also showed that the nature of the blob motion depends on the Ca , the viscosity ratio, and the water saturation. Larson *et al.* (1977) showed that for a given pore space morphology, the critical length of a blob blew which it is trapped is proportional to Ca^{-1} .

Thus, mobilization and displacement of trapped oil blobs require relatively high capillary numbers, say $Ca > 10^{-4}$. The value of the required Ca depends, however, on several factors, including the shape and size of the blobs, the morphology of the porous medium, especially around the regions where the blobs reside, and the contact angle. For $Ca > Ca_c$, where Ca_c is a critical value of Ca , the blobs begin to move. If $Ca - Ca_c$ is small, then we obtain what is called *quasi-static displacement* of the blobs, during which one blob moves downstream, while one or two may move upstream. A blob may get re-entrapped if it arrives at a pore body where all the throats that are connected to it are too small for the blob movement, in which case one needs an even higher Ca to move such blobs.

A moving blob is almost certain to break into smaller blobs by one of the following mechanisms. In *pinch-off*, the velocity of the moving blob becomes small for a long enough time that the blob collapses into several smaller ones. In *dynamic breakup* (Payatakes, 1982), a blob advances in two or more pore throats simultaneously, which can easily happen if the coordination number of the pore space is large enough. For this to happen, the Ca must be large enough that even if two pore throats connected to the same pore body have different effective sizes, there can still be enough force to move the blob into both pores. If the blob completely

evacuates the pore body, it breaks into two or more smaller blobs, depending on how many pore throats it enters.

There is yet another mechanism for blob breakup that is usually called *choke-off* or *snap-off*, and was first discussed by Pickell *et al.* (1966). Roof (1970), Mohanty *et al.* (1980, 1987), and Arriola *et al.* (1983) all described this phenomenon in detail. Choke-off signals the breakup of a small drop from the leading tip of a non-wetting thread that tries to pass through a narrow constriction. Roof (1970) conducted several experiments and demonstrated the phenomenon nicely. He considered choke-off in a toroidal pore throat, assumed that there is a thin lubrication film on the surface, and showed that choke-off occurs if the curvature of the interface at the throat is larger than the curvature of the tip of the thread. If the throat is non-axisymmetric, then Roof showed that the same phenomenon happens, except that it takes place faster than the toroidal case because the film has easier access to the point of rupture as a result of the fact that some parts of the cross section are not filled by the non-wetting film. Roof also showed that before choke-off occurs, the tip of the non-wetting fluid thread must travel a distance of several pore-throat diameters beyond the original throat. Therefore, choke-off is important if the ratio of the pore body and pore throat diameters is large. Roof's prediction was demonstrated by the experiments of Wardlaw (1982) and Li and Wardlaw (1986a,b). Based on their detailed experimental observations, Chatzis *et al.* (1983) determined that roughly 80% of trapping of the non-wetting phase occurs by snap-off geometries, with the rest happening in pore doublets or a combination of both pore geometries (see Figure 14.7).

Hammond (1983) made a careful study of this phenomenon by solving the problem of slow adjustment of lubricated threads and drops in axisymmetric, straight capillaries, and constricted tubes. He showed that for choke-off to happen, it is not sufficient for the lubrication film to be unstable. One must also have a sufficient amount of wetting liquid in the film near the incipient neck to form a bridge across

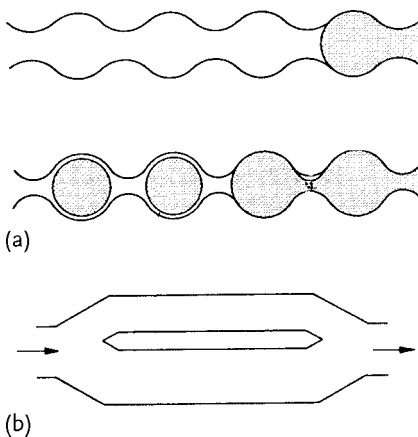


Figure 14.7 Snap-off geometries (a) and the pore doublet model (b). Dotted areas represent the oil phase, while the white area is the water phase (after Chatzis *et al.*, 1983).

the tube because while the neck is being formed, the thread on either side bulges and isolates the local wetting fluid from the rest. Hammond estimated that for the rupture to occur, the thickness ℓ_w of the wetting fluid, normalized by the tube radius, must be larger than $\pi/6$. He also estimated that the time scale for the growth of a perturbation large enough to cause the rupture of a thread in a constriction is at least of the order $\ell_w^{-2} D \mu_1 / \sigma_{fs}$, where D is the diameter of the tube.

A simple criterion for snap-off can be derived using the observations described. We assume that the non-wetting phase flows through a single path of variable cross-sectional area. The sides of the flow path are lubricated by the wetting fluid so that it is possible to define a local capillary pressure everywhere along the path that may, however, vary from point to point. For example, if the path is narrow, then the local capillary pressure is large. Depending on the potential gradient and the geometry of the path, the capillary pressure across the path can be larger than the potential gradient across the same segment of the path. If so, the external force would not be large enough to force the non-wetting phase to enter the next pore segment, in which case the non-wetting fluid snaps off into globules that reside in the pore bodies along the path. If such a description is accurate, then the condition for mobilizing the trapped globules is given by

$$\Delta \Phi + g \ell_g \Delta \rho \sin \alpha_g \geq \Delta P_c, \quad (14.15)$$

where $\Delta \Phi$ and ΔP_c are the wetting phase potential and capillary pressure changes across the globule, ℓ_g is the size of the globule, $\Delta \rho$ is the difference between the densities of the two fluids, and α_g is the angle between the globule's major axis and the horizontal axis. Based on inequality Eq. (14.16) and the work of Melrose and Brandner (1974), Stegemeier (1974) derived the following condition for mobilization of a trapped globule

$$Ca_1 = \frac{K_e |\nabla P_w|}{\sigma_{wnw}} \geq \frac{\phi}{c} \left[J \cos \theta_A - \frac{\cos \theta_R}{\sqrt{2\tau}} \right]^2. \quad (14.16)$$

Here, Ca_1 is a local capillary number, ∇P_w is the pressure gradient in the wetting phase, σ_{wnw} is the interfacial tension between the wetting and non-wetting phases, ϕ is the porosity of the medium, J is the Leverett J -function (see Eq. (4.27)), θ_A and θ_R are the advancing and receding contact angles, τ is the tortuosity factor of the porous medium, and c is a constant of about 20. Observe that Eq. (14.16) expresses the condition for mobilizing the non-wetting fluid globule in terms of measurable or easily estimated quantities. Moreover, it allows the effect of contact angles to be included in the condition, thus making it more realistic.

Finally, just as blobs breakup into several smaller blobs, they can also coalesce, which happens when two interfaces that belong to two different blobs pass through the same throat and are pressed against each other for a long enough time. Constantinides and Payatakes (1991) used a pore network model to investigate the likelihood of collision and coalescence of blobs in a porous medium. Their results indicated that the wetting characteristics are more important than Ca , M , or θ , and that the probability of coalescence, given a collision, decreases as θ increases.

They estimated the probability to vary between 0.03 and 0.15. Thus, the breakup and coalescence phenomena give rise to a series of complex dynamical processes in which the displaced fluid can break, but form a large cluster again at a later time. Lenormand and Zarcone (1985b) presented nice experimental realizations of such phenomena in a micromodel.

14.10

Relative Permeability

Just as slow single-phase flow through a porous medium is quantified by the effective permeability K_e , so also are multiphase flows. Hence, for two fluid phases β and γ , one has *phase* permeabilities K_β and K_γ . One must first generalize the Darcy's law to multiphase flow, in order to obtain a better understanding of how the phase permeabilities arise. Such a generalization has been developed by many. In particular, Whitaker (1986b) studied flow of two immiscible fluids in porous media by starting with the continuity and Stokes' equations for each phase, and using the appropriate boundary and initial conditions. He derived the following equations for the average flow velocity and saturations of the fluid phases β and γ ,

$$\mathbf{V}_\beta = -\frac{\mathbf{K}_\beta}{\mu_\beta} \cdot (\nabla(P_\beta)^\beta - \rho_\beta \mathbf{g}) + \mathbf{K}_{\beta\gamma} \cdot \mathbf{V}_\gamma, \quad (14.17)$$

$$\phi \frac{\partial S_\beta}{\partial t} + \nabla \cdot \mathbf{v}_\beta = 0, \quad (14.18)$$

$$\mathbf{V}_\gamma = -\frac{\mathbf{K}_\gamma}{\mu_\gamma} \cdot (\nabla(P_\gamma)^\gamma - \rho_\gamma \mathbf{g}) + \mathbf{K}_{\gamma\beta} \cdot \mathbf{V}_\beta, \quad (14.19)$$

$$\phi \frac{\partial S_\gamma}{\partial t} + \nabla \cdot \mathbf{v}_\gamma = 0, \quad (14.20)$$

where all notations are as before. Note that Eqs. (14.17) and (14.19) contain two terms, the first of which is the usual Darcy's law written for each phase, while the second term is a cross term that couples the two phases. Equations (14.17) and (14.19) were first proposed by Raats and Klute (1968) and de Gennes (1983b) based on physical arguments, although somewhat similar equations had been conjectured by Rose (1972).

In analogy with the thermodynamics of irreversible processes, one may assume that $\mathbf{K}_{\beta\gamma} = \mathbf{K}_{\gamma\beta}$. Equations (14.17) and (14.19) are valid if the capillary number $\text{Ca} \ll 1$, and if moving contact lines do not have a significant effect. Generally speaking, the coupling terms in Eqs. (14.17) and (14.19) are not significant unless $\mu_\beta \simeq \mu_\gamma$ (in which case a thin film of one fluid phase coats the walls of a pore the bulk volume of which is filled with the other phase). Some of the most convincing evidence for the insignificance of the cross terms of Eqs. (14.17) and (14.19) was provided by Yadav *et al.* (1987). They experimented with a wetting and a non-wetting fluid and showed that the permeability of both phases in drainage, measured when the opposite phase was solidified in situ, was the same as that typically

measured for Berea sandstones. However, Kalaydjian and Legait (1987) and Goode and Ramakrishnan (1993) showed that such coupling terms might be important in certain cases, even if μ_β and μ_γ are not close to each other.

In practice, one calculates the phase permeabilities by the use of the relations

$$K_\beta = K_e k_{r\beta} , \quad (14.21)$$

with a similar equation for the γ -phase, where $k_{r\beta}$ is called the *relative permeability* (RP) to the β -phase, a concept that has been used for many decades in the petroleum industry. A major problem in multiphase flows in porous media is the prediction of the RPs. Unlike the absolute permeability K_e , $k_{r\beta}$ has been found to depend on many parameters, including saturation and saturation histories of the fluids (Johnson *et al.*, 1959; Naar *et al.*, 1962), pore space morphology (Morgan and Gordon, 1970), the wetting characteristics of the fluids (Owens and Archer, 1971; McCaffery and Bennion, 1974), sometimes, on the viscosity ratio (Odeh, 1959; Le Febvre du Prey, 1973), and the capillary number Ca (Leverett, 1939; Taber, 1969). Moreover, nearly 60 years ago it was recognized (Richardson *et al.*, 1952) that the RP to a fluid phase typically becomes small or altogether negligible when its saturation is less than a critical value that is distinctly larger than zero. The existence of such critical saturations, is, of course, the signature of a percolation problem. Thus, various percolation models have been developed for modeling two-phase flows in porous media that will be described in Chapter 15. The reason $k_{r\beta}$ apparently depends on the saturation history of a fluid phase, that is, the way a saturation has been reached, is that there are presumably multiple shapes of the pore regions that satisfy the Stokes' equation which is made nonlinear by the free interfaces. The saturation history dependence naturally gives rise to *hysteresis* in the RPs, which will be described shortly.

14.11

Measurement of Relative Permeabilities

Similar to most properties of fluid flow in porous media, the RPs may also be measured by a variety of methods. The most direct and the least controversial method for measuring the RP is the steady-state method in which the total flow rates of oil and water are usually kept constant, while their ratio is varied at the inlet of the core. After a change, it is necessary to wait until steady state is reached in the core. The individual flow rate and pressure drop in each fluid phase are then used to calculate the individual phase RP by using the Darcy's law. The core average saturation must be measured independently in order to determine its relationship with the RP.

The main problem with the steady-state method is the basic assumption that the capillary pressure can be neglected. Actually, due to the capillary effects, the saturation distribution along the core is nonuniform, and the pressure drop is not the same in both fluid phases. One must also deal with the *end effects*, which refer to a discontinuity in the capillary properties of the porous medium at the effluent

end of the core. Within the core, capillary forces act uniformly in all directions, thus canceling each other. Though at the outlet, a net force persists that tends to prevent the wetting phase from leaving the core. Therefore, the wetting phase saturation S_w must build up near the boundary of the medium, which can cause serious error in the RP measurements if the core average saturations and pressure drops are used for the RP estimation. For reliable measurements, it is necessary to either minimize the end effects or to account for them in the calculation of the RP. Several methods have been reported in the literature for this purpose, which are reviewed by Honarpour *et al.* (1986). In what follows, we describe a few steady-state methods.

14.11.1

The Hassler Method

The capillary pressure or the Hassler method (Hassler, 1944) is based on the idea that the end effects will not arise if the fluids do not have to leave the porous medium simultaneously from the same location. To ensure this condition, semipermeable membranes are installed at each end to produce different fluids from different outlets. Using the same technique for measuring the RPS for three-phase flow (water, oil, and gas) requires three semipermeable membranes, with each one being permeable to one fluid only. If only two membranes, one permeable to water and another to oil and gas, are used, the end effects would be reduced but not completely eliminated. The method is quite cumbersome and is currently not very popular.

14.11.2

The Penn-State Method

One of the most commonly used method is perhaps the Penn-State method which uses separate inlet and outlet sections of cores with similar material. The saturation and pressure drop are measured in the central piece only, which represents the core to be tested. The method should work reasonably well for three-phase flows also, provided that a perfect capillary contact can be ensured between the adjoining cores. Such a method of minimizing the end effects makes the saturation measurement somewhat more difficult. Unless in-situ techniques are used, the saturation measurement requires removing the test core from the core holder for each measurement.

14.11.3

The Richardson–Perkins Method

An interesting technique which has received very little attention was suggested by Richardson and Perkins (1957). Their method relies on attaching a narrow packed tube to the outflow end of the core. Due to a very high flow rate in the narrow section, the end effects remain confined to the narrow tube section. A short narrow section must be used to minimize the effect of saturation variation in this section on the core average saturation measurements by material balance. Its improve-

ment, based on multirate steady-state techniques, was proposed by Virnovsky *et al.* (1998). For a fixed fractional flow at the inlet, a number of steady-state experiments is required with varying total flow rates in order to include the capillary effects in the analysis of the data. The capillary pressure curve must be measured separately, or determined simultaneously (Urkedal *et al.*, 2000). All parts of the core must follow the same hysteresis curve for both primary drainage and primary imbibition.

Although more time consuming and notably more expensive compared to the unsteady-state methods (see below), the steady-state techniques are usually recommended (Crotti and Rosbaco, 1998) for the following reasons:

1. The results are unaffected by the heterogeneities of porous media. In the case of stratified media, after reaching the stationary state, no intercross flow occurs and the RPs measured represent an average of RP for each layer. The unsteady-state methods (see below) produce transient states that give rise to the “anomalous” RP curves.
2. The calculations are very simple.

However, the following aspects can sometimes make the steady-state methods questionable.

1. The use of long measurement times and large injection volumes may affect the results due to fluid interaction with the porous medium.
2. In homogeneous media, measurements using the unsteady-state methods lead to the same results as those obtained with the steady-state methods. This is not, however, true for heterogeneous porous media, for which the steady-state method eliminates the influence of heterogeneities.
3. If the samples are heterogeneous, the heterogeneity will be present in the reservoir’s bed and, therefore, the curves characteristic of homogeneous media lack any representativity.
4. Stationary conditions do not usually occur in a large-scale porous medium such as an oil reservoir.
5. If viscous forces are not predominant in the porous medium, only the end points of the RP curves are useful for the reservoir engineer.

14.11.4

Unsteady-State Methods

For a discussion of the differences between the steady-state and unsteady-state RPs, see Alemán *et al.* (1989). Measurement of the RP by an unsteady-state method is the most commonly used method because it is fast and qualitatively resembles the flooding process (of oil or water) in oil fields. The RPs are calculated, not measured, which makes the material balance analysis more difficult than the steady-state methods. In an unsteady-state method, one or two fluid phases are injected into a core saturated initially with other fluid(s). Then, effluent composition and the pres-

sure drop across the core are monitored. The RP values of the in-situ and the driving fluid(s) are then calculated using an equation originally developed by Buckley and Leverett (1942), to be described in the next section. Examples of the unsteady-state methods are the experiments of Johnson *et al.* (1952), Welge (1959), and Jones and Roszelle (1978). Such methods are based on the following assumptions:

1. capillary pressure effects are negligible compared with viscous effects.
2. The core is a linear, homogeneous body.
3. The flow of the fluids is stable and one-dimensional.

In practice, such assumptions are often not met. Capillary forces, viscous fingering (see Chapter 13), and heterogeneity influence the flow in a core plug and, hence, the RP (Mohanty and Miller, 1991; Ringrose *et al.*, 1996). Because the point pressure gradient per unit injection rate is required by the JBN method in order to calculate the RPs, it involves conversion of the average injectivity to a point value that requires conversion of experimental data involving complex computations and evaluation of the derivatives. Hence, the computations may introduce serious errors into the analysis.

In order to circumvent such errors, Jones and Roszelle (1978) proposed a graphical technique, which could be applied to constant rate, constant pressure as well as variable rate and pressure displacements. In both methods – the JBN method and that of Jones and Roszelle – inaccuracies in the data measurement are amplified by the process of differentiation. Another major drawback of such methods is that they use the breakthrough point as a very important parameter. As a consequence, any error in estimating that point may yield erroneous results. However, with the exception of a minor difference with regard to the base permeability, the two methods are essentially similar. In addition to the aforementioned methods and their improved versions, for example, those proposed by Sigmund and McCaffery (1979), Marle (1981), Ruth *et al.* (1988), and Toth *et al.* (2001), which are applicable for high-rate displacement tests in which the end effects may be negligible, various methods have also been developed for low-rate displacements in which the capillary pressure may be significant (Islam and Bentsen, 1986; Kalbus and Christiansen, 1995; Helset *et al.*, 1998; Goodfield *et al.*, 2001).

Many researchers have reported disagreement between the RPs measured by the unsteady-state and steady-state methods. For example, Aleman-Gomez *et al.* (1984) concluded from their theoretical study of unsteady-state displacement that the two methods are similar only under certain limited conditions. They also stated that if the discrepancy between the steady-state and unsteady-state RPs is not observed, it is most likely due to the fact that the latter method is not capable of describing the truly unsteady-state flow. A hint for the existence of this problem was already provided by the early work of Kimbler and Caudle (1957), who concluded that the flow distributions for the steady-state condition differ significantly from what pertains to unsteady-state flow. Unfortunately, the conventional JBN-type methods estimate the RPs at saturations corresponding to those pertaining at the outlet face of a 1D flow system. However, as Aleman-Gomez *et al.* (1984) pointed out, the dis-

placement front coincides with the outlet face only at one point in time. They also showed that the RPs measured by the JBN method correspond to the small values of *local* capillary number and, thus, are indistinguishable from those measured by the steady-state methods, unless the core is subjected to an unusually large pressure gradient.

Heaviside *et al.* (1983) and Crotti and Rosbaco (1998) described some of the shortcomings of the unsteady-state methods:

1. The RP data will not be over the entire saturation range, and might only be restricted to the end point data.
2. Discontinuities in the capillary pressure at the outlet and the forces at the core outlet may lead to distortion of the recovery and pressure response data that are measured.
3. Variations of data with the rate and length may also be observed, again due to capillary influences.
4. Most of the RP estimations are performed under the conditions that correspond to steady-state flow (homogeneous saturation distribution), whereas the intermediate values of the RPs are calculated during a transient stage in which the sample saturation is not homogeneous. As a result, the end point saturations should have greater validity than the remaining points in the curve since the shape of the RP curves in heterogeneous systems depends – among other things – on the mobility ratio, whereas it is generally accepted that the end points – if properly extrapolated at the end of the test – are independent of it.
5. It is generally believed that residual oil saturation, S_{ro} , is obtained only after injecting “infinite” pore volumes of water. S_{ro} can, however, also be attained through capillary/gravitational equilibrium that depends solely on time. In a large-scale porous medium, such as, an oil reservoir, the times involved are much longer than those employed in the lab.
6. Although there are different ways of reaching the end points, as stated before, they tend to be numerically different, depending on whether they are provided by capillary pressure tests or from the RP tests.
7. Measurement of the end point saturations depends on the interpretations and assumptions of the user. In the case of S_{ro} , the experimenter must extrapolate the production data until “infinite” pore volumes of water are injected. Such extrapolation is usually not valid for heterogeneous porous media where the production curves exhibit inflections derived from the different productivity and response time of different layers. For this reason, extrapolation leads to different values for S_{ro} , depending on the time selected for ending the flooding. As is well known, there is currently no universal criterion for making the decision of when to end the test.
8. The RP curves obtained by laboratory experiments only include the effect of the viscous forces since the measurements and computations are performed in such a way that minimizes the contribution of the capillary and gravitational forces. However, as emphasized by Dake (1994), capillary as well as gravitational forces are often the predominant forces at the reservoir scale.

Also of importance is the requirement that the model and the prototype be *dynamically* similar. Such similarity in fluid displacement studies is achieved by requiring that the ratios of the viscous forces of the gravitational forces to the viscous forces and of the capillary number Ca have the same values in the model and the prototype. Model studies of the kind described so far implicitly assume that the flow regime is the same in the model and the prototype. Moreover, the implicit assumption is that Darcy's law is satisfied in both systems, but it is important to recognize the existence of such implicit assumptions.

During the early stages of a displacement experiment, the fraction of water flowing at a particular location within the core is dependent upon not only the water saturation, but also on the amount of water that is being injected. As a consequence, the shape of the frontal region of the saturation profile is time dependent. As water injection continues, the shape of the frontal region eventually becomes invariant with time, at which point the displacement is said to be stabilized. As stabilization is approached, the shapes of the saturation profiles approximate those predicted by Buckley–Leverett theory (see the next section). Dimensional analysis suggests that the amount of water that must be injected in order to achieve stabilization is directly proportional to the linear capillary number Ca and inversely proportional to the end point mobility ratio M_e .

14.11.5

Relative Permeabilities from Capillary Pressure Data

Although estimating the RPs based on capillary pressure data is not usually the preferred technique, such methods are useful when the samples are too small for flow tests, but are large enough for mercury injection. The techniques are also useful for porous media that have very low permeability as well as for the case in which the capillary pressure data have already been measured, but a sample is not available for measuring the RPs. Porous media with special fluids, such as, gas condensate and geothermal reservoirs in which there are phase transformation and mass transfer between the two phases, also represent the cases for which the use of the P_c data for estimating the RPs is useful.

Li and Horne (2003) demonstrated that the uncertainty in core and reservoir simulations may be reduced if the number of input parameters is decreased, especially if the parameters with the greatest uncertainty are avoided. This may be realized by only using capillary pressure data as the input to numerical simulators, and inferring the RPs by correlating them with the capillary pressure. Purcell (1949), Corey (1954), and Brooks and Corey (1966) represented the capillary pressure curves as power-law functions of S_w , which have been used widely. Honarpour *et al.* (1986) reviewed the literature on the empirical correlation between the RP and capillary pressure in drainage. Skjaeveland *et al.* (2000) developed a general capillary pressure correlation for primary drainage, imbibition, secondary drainage and history scanning loops. The correlation composed as a sum of two terms, each with two adjustable parameters. Papatzacos and Skjaeveland (2002) developed a theory for single-component, two-phase flow in porous media that included the effects of wet-

tability and capillary pressure as integral parts of the thermodynamic description, and did not make use of the RP. Li and Horne (2002) demonstrated that the RPs for two-phase flow in many porous media is calculated satisfactorily once reliable capillary pressure data are available.

There are many advantages to inferring the RPs from capillary pressures data and/or models. Measurements of the RPs over the full range of the saturation are usually time-consuming, expensive, and inaccurate in many cases, whereas measurements of P_c are faster, cheaper, and more accurate. At the same time, the correlation between P_c and rock properties is established experimentally much better than that between the RP and the same properties. Unlike the commonly used method, namely, tuning the RP curves, one may match the production history of an oil reservoir by tuning the capillary pressure curves that have physical significance based on the well-established correlation between capillary pressure and rock properties, that is, the Leverett J -function. Another advantage is that the uncertainty in the results of any numerical simulations may be reduced because the number of input parameters is decreased.

The Brooks–Corey model (Brooks and Corey, 1966), which uses capillary pressure data to estimate the RPs, has been used widely. In many cases, Purcell’s empirical model has been, however, found to provide the most accurate fit to the experimental data for the wetting-phase RP, with the differences between the experimental and the model data being almost negligible. Li and Horne (2002) concluded that the wetting-phase RP can be calculated using the Purcell model, while the nonwetting-phase RP can be estimated using the Brooks–Corey model. According to them, the wetting-phase RP can be calculated accurately using the following equation:

$$k_{rw} = (S_w^*)^{\frac{2+\lambda}{\lambda}} \quad (14.22)$$

where S_w^* is the normalized saturation of the wetting phase (see below), and λ is a parameter related to pore size distribution of the sample. Equation (14.22) was derived by substituting the following Brooks–Corey capillary pressure model into the Purcell model,

$$P_c = P_e (S_w^*)^{-\frac{1}{\lambda}} \quad (14.23)$$

where P_e is the entry capillary pressure. The normalized saturation of the wetting phase in drainage is given by

$$S_w^* = \frac{S_w - S_{rw}}{1 - S_{rw}} \quad (14.24)$$

where S_w and S_{rw} are the saturation and the residual saturation of the wetting phase. For the non-wetting phase, the RP can be calculated accurately using the Brooks–Corey model,

$$k_{rnw} = (1 - S_w^*)^2 \left[1 - (S_w^*)^{\frac{2+\lambda}{\lambda}} \right]. \quad (14.25)$$

We emphasize that such correlations are empirical, but useful.

14.11.6

Relative Permeability from Centrifuge Data

The flow process in a centrifuge is similar to core flood under unsteady-state condition with prescribed inlet and outlet pressures. Hagoort (1980) neglected the capillary pressure and mobility ratio terms in the governing flow equation (see below) in order to derive an analytical solution that can be used for estimating the oil-phase RP in a gas-oil system. His method was extended by van Spronsen (1982) and O'Meara and Leas (1983) that enables one to estimate the oil and water phase permeabilities K_w and K_o . Firoozabadi and Aziz (1986) applied various RP models to the centrifuge experimental data to show that (1) recoveries (of oil in a displacement experiment) can be matched with two – and perhaps even more – very different sets of RP curves, and (2) good history match (matching of production of data for an oil reservoir with the simulated data) is possible with arbitrarily small capillary pressures, but the resulting RPs will be different from those obtained based on the experimental data for the capillary pressure. Thus, the capillary pressure term should not be ignored in obtaining the RPs from centrifuge data (see also Skauge and Poulsen, 2000).

In a survey by the Society of Core Analysis (www.SCAweb.org) on drainage capillary pressure measurements using centrifuge of many samples of Berea sandstone and Bedford limestone, significant differences between the data were identified, even though the samples were all comparable in terms of their porosity and single-phase permeability. In fact, in the centrifuge method, the capillary pressure curve is not directly measured, but is calculated from fluid production measurements, using various approximate methods (see Chapter 4). The survey demonstrated that the main source of inaccuracy is related to the interpretation process, and not to the experimental procedures nor to the shape of the capillary pressure curve, and that the inaccuracy depends on the centrifuge geometry (and the resulting contributions of centrifugal and radial forces), and on the method used for solving the centrifuge equation.

14.11.7

Simultaneous Estimation of Relative Permeability and Capillary Pressure

Newly developed steady-state methods compensate for the capillary end effects by including the capillary term in the governing equations (see the next section), but require measurement of the pressure in each phase and, thus, are as time consuming as the traditional steady-state methods. For both the unsteady-state and steady-state methods, the capillary pressure must be determined by independent experiments which requires multiple experiments on the same core, hence necessitating re-establishment of the same initial states and wetting conditions. Such a procedure is both difficult and time consuming. Measurement of fluid saturations within the core samples yields significant information about the flow functions. Kulkarni *et al.* (1996) used nuclear-magnetic resonance imaging (see Chapters 4, 10, and 11), and Honarpour *et al.* (1996) utilized microwave signals to obtain sat-

uration profiles that were then used for estimating the multiphase flow functions from unsteady-state and steady-state methods, respectively. See also Ohen *et al.* (1991) and Ucan and Cinar (1996).

14.12

Effect of Wettability on Relative Permeability

We already described the effect of wettability on capillary pressure. It should be clear that wettability and contact angles also affect the RPs. Figure 14.8 presents typical oil-water RP curves for a strongly water-wet system, and for a strongly oil-wet porous medium. While the difference between the two RP curves for the oil phase is not very large, there is a dramatic difference between the RPs to the water phase. Note also the existence of a finite saturation (i.e., a percolation threshold) at which the RP vanishes. Normally, if a porous medium is strongly water-wet, there is little or no hysteresis in the RPs to the water phase. This can be clearly seen in the experimental data of Morrow and McCaffery (1978). They measured the RPs in a teflon core with nitrogen as the non-wetting fluid and heptane ($\theta = 20^\circ$) and dodecane ($\theta = 42^\circ$) as the wetting phase and found that there is no hysteresis in the wetting-phase RP.

Figure 14.9 shows the data of McCaffery and Bennion (1974) and Morrow and McCaffery (1978) for the three different wettability regimes described above, namely, wetted, intermediately-wetted, and non-wetted cases. Here, the reference phase (with saturation S_r) refers to the displaced phase, while the non-reference phase refers to the displacing phase. The differences between the three cases are rather large. For example, in the non-wetting case, the contact angle for the non-reference phase was more than 130° , whereas in the wetted case, it was at most 49° . It is

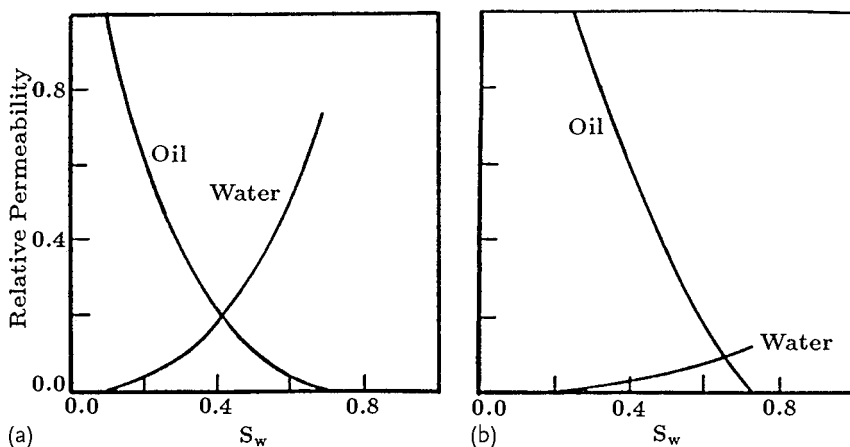


Figure 14.8 Typical relative permeability curves for an oil-wet (a) and a water-wet (b) porous medium (after Craig, 1971).

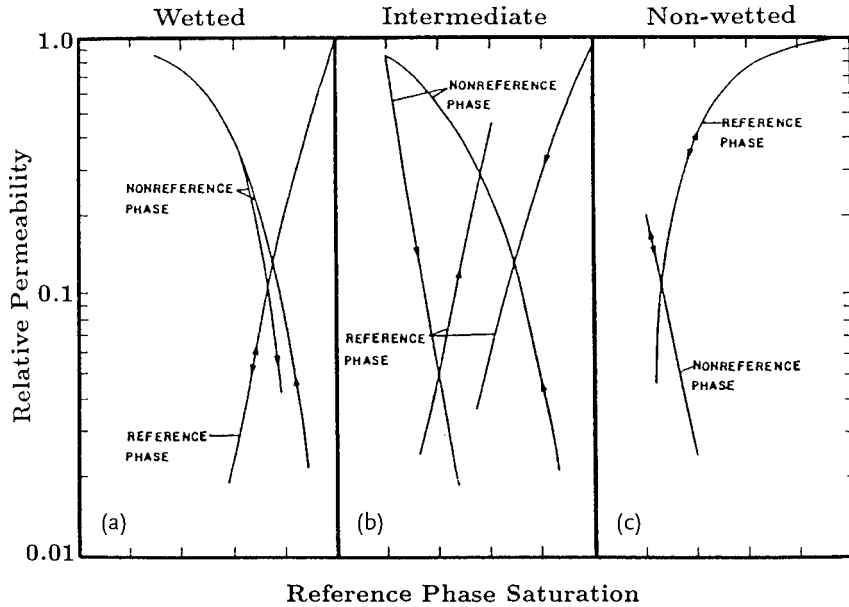


Figure 14.9 Relative permeability versus the reference phase saturation S_r for various wettability regimes (a–c). The contact angles for the wetted (a) and non-wetted (c) cases were 49° and 130° , respectively (after McCaffery and Bennion, 1974 and Morrow and McCaffery, 1978).

clear that wettability strongly affects the trends in RPs, and one major theoretical challenge is to predict such trends. We will come back to this important issue in Chapter 15.

14.13

Models of Multiphase Flow and Displacement

Similar to all the phenomena described so far in this book, there are two classes of models of multiphase flow and displacement in porous media. One of them relies on the continuum equations, averaged over suitably defined representative elementary volume. The approach represents the classical engineering approach, the major elements and achievements of which were already described in the previous chapters. The literature on this class of model is enormous, and except for its most important aspects, there is no possibility of describing it in this chapter. We refer the reader to Collins (1961), Bear (1972), Scheidegger (1974), and particularly Marle (1981) for comprehensive discussions of this class of models. Another class of approaches to multiphase flow in porous media utilizes the pore network models already described in the previous chapters for single-phase, transport and dispersion, and miscible displacements. Over the past 30 years, the literature on this class of models has also grown enormously. We will describe the most important

and fruitful of such models in Chapter 15. For now, let us describe a classic continuum model of two-phase flows in porous media, namely, the celebrated Buckley–Leverett model.

14.14

Fractional Flows and the Buckley–Leverett Equation

We consider a 1D displacement and ignore the cross terms in Eqs. (14.17) and (14.19). We also assume that there are only two fluid phases in the porous medium. If the displacement is along the z direction, then Eqs. (14.17)–(14.20) become

$$V_w = -\frac{K_e k_{rw}}{\mu_w} \left(\frac{\partial P_w}{\partial z} - \rho_w g \right), \quad (14.26)$$

$$V_{nw} = -\frac{K_e k_{rnw}}{\mu_{nw}} \left(\frac{\partial P_{nw}}{\partial z} - \rho_{nw} g \right), \quad (14.27)$$

$$\phi \frac{\partial S_w}{\partial t} + \frac{\partial V_w}{\partial z} = 0, \quad (14.28)$$

$$\phi \frac{\partial S_{nw}}{\partial t} + \frac{\partial V_{nw}}{\partial z} = 0. \quad (14.29)$$

Moreover, we also have $S_w + S_{nw} = 1$ and $P_{nw} - P_w = P_c$. We now define new variables f_w and f_{nw} , called the *fractional flows*, by

$$f_w = \frac{V_w}{V}, \quad (14.30)$$

$$f_{nw} = \frac{V_{nw}}{V} = 1 - f_w, \quad (14.31)$$

where $V = V_w + V_{nw}$. It is not difficult to show that

$$f_w = \frac{\frac{\mu_{nw}}{k_{rnw}} + \frac{K(\rho_w - \rho_{nw})g}{V}}{\frac{\mu_w}{k_{rw}} + \frac{\mu_{nw}}{k_{rnw}}} + \frac{K_e}{V} \frac{1}{\frac{\mu_w}{k_{rw}} + \frac{\mu_{nw}}{k_{rnw}}} \frac{\partial P_c}{\partial z}. \quad (14.32)$$

If the capillary pressure gradient is negligible, $\partial P_c / \partial z \simeq 0$, which is the case if the flow rate or the fluid velocity is large, and if the density difference $\rho_w - \rho_{nw}$ is also small (or the gravity is not important), Eq. (14.32) is simplified to

$$f_w = \left(1 + \frac{K_{nw}}{K_w} \frac{\mu_w}{\mu_{nw}} \right)^{-1} = \left(1 + \frac{k_{rnw}}{k_{rw}} \frac{\mu_w}{\mu_{nw}} \right)^{-1}. \quad (14.33)$$

We now use the relation $\partial f_w / \partial z = (d f_w / d S_w)(\partial S_w / \partial z)$ to rewrite the continuity equation for the wetting phase, Eq. (14.28), in the following form,

$$\left(\frac{V}{\phi} \frac{d f_w}{d S_w} \right) \frac{\partial S_w}{\partial z} = -\frac{\partial S_w}{\partial t}, \quad (14.34)$$

with a similar equation for the non-wetting phase. Equation (14.34) is nonlinear, as $d f_w / d S_w$ depends on S_w and, therefore, does not, in general, have an explicit analytical solution. On the other hand, we can write

$$\frac{d S_w}{d t} = \frac{\partial S_w}{\partial z} \frac{d z}{d t} + \frac{\partial S_w}{\partial t}, \quad (14.35)$$

so that if $z = z(t)$ is selected to coincide with a surface of fixed S_w , we have $d S_w / d t = 0$, and

$$\left(\frac{d z}{d t} \right)_{S_w} = - \frac{\partial S_w}{\partial t} \left(\frac{\partial S_w}{\partial z} \right)^{-1}, \quad (14.36)$$

which, together with Eq. (14.34), yields

$$\left(\frac{d z}{d t} \right)_{S_w} = \frac{V}{\phi} \frac{d f_w}{d S_w}. \quad (14.37)$$

Equation (14.37) is the celebrated Buckley–Leverett equation (Buckley and Leverett, 1942), widely used in the petroleum industry. We should emphasize that the Buckley–Leverett equation is valid when the gradient of the capillary pressure and the density difference between the two fluids (or gravity) are negligible. Moreover, we should keep in mind that because f_w depends on the phase permeabilities or the RPs, it also depends on the phase saturation S_w and, hence, $S_{nw} = 1 - S_w$. Integrating the Buckley–Leverett equation with respect to time, we obtain

$$z(t) - z(0) = \frac{V(t) - V(0)}{\phi} \frac{d f_w}{d S_w}. \quad (14.38)$$

Consider, as an example, the RP curves shown in Figure 14.8. The curves are used to construct the fractional flow curves if the ratio of the viscosities of the two fluids is known. Consider the simplest case in which the viscosity ratio is unity. Then, given the RP curves, the corresponding fractional flow curve and its derivative $d f_w / d S_w$ are shown in Figure 14.10. Suppose that the initial saturation profile consists of all saturations $S_c < S_w < 1$ that exist at $z = 0$ and $t = 0$, where S_c is the *connate* or *irreducible* wetting-phase saturation. If we use Eq. (14.38) to construct the saturation profile at time t , we obtain the results that are in Figure 14.11, indicating that one may obtain multiple values of the saturation, an unphysical situation. To remove the multiplicity, we write a volumetric balance for the wetting phase

$$Q(t) = \int_0^{z_c} A \phi (S_w - S_c) d z, \quad (14.39)$$

where A is the cross-sectional area of the system. Here, z_c is a cutoff point beyond which $S_w = S_c$, which introduces a discontinuity in the saturation profile. If we

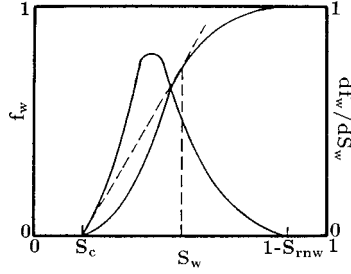


Figure 14.10 Wetting-phase fractional flow f_w and its derivative versus the wetting-phase saturation S_w , constructed based on the relative permeabilities shown in Figure 14.8a.

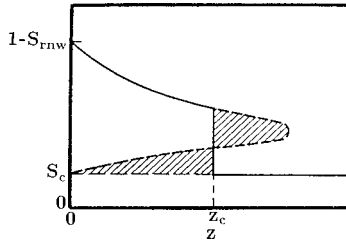


Figure 14.11 The Buckley-Leverett prediction of the distribution of the wetting-phase saturation, indicating the discontinuity.

let S_{cw} be the upper saturation at the discontinuity, use $Q(0) = 0$, $z(0) = 0$, and $f_w = 0$ at $S_w = 1 - S_{rnw}$, and carry out the integration by parts, we obtain

$$\frac{d f_w(S_{cw})}{d S_w} = \frac{f_w(S_{cw})}{S_{cw} - S_c}. \quad (14.40)$$

Equation (14.40) indicates that the multiplicity in the saturation profile can be removed, if we remove all the saturations below S_{cw} , which is a particular saturation evaluated as the saturation at the point at which a straight line passing through the point $f_w = 0$ and $S_w = S_c$ is tangent to the curve $f_w(S_w)$.

The existence of such multiplicities makes the analysis of immiscible flows difficult. Moreover, use of Eq. (14.28) in Eq. (14.32) leads to a second-order nonlinear equation for S_w for the general problem of flow of two immiscible fluids in a porous medium that can be solved mostly by numerical simulations. Marle (1981) provides a good qualitative discussion of this problem, to whom the interested reader is referred.

14.15

The Hilfer Formulation: Questioning the Macroscopic Capillary Pressure

As the discussions in Chapter 4 and the present chapter clarify, the notion of a macroscopic capillary pressure is the foundation of the continuum theories of mul-

tiphase flow in porous media. Hilfer (2006a,b,c), however, challenges this view. The basis of his challenge is the physically appealing argument that one must distinguish between the percolating and non-percolating fluid regions (Hilfer, 1998). Hence, Hilfer develops a new formulation, starting from Eqs. (14.18) and (14.20), but *without* supplying the capillary pressure as an input of the model. The saturations of the flowing and non-flowing fluids are treated separately. The capillary pressure, an *output* of the theory, depends not only on the water saturation – as in the traditional theories described earlier – but also on the saturations of the non-flowing parts of the fluids. The RPs are also no longer the inputs to the theory, but rather the output. The model does involve adjustable parameters that, however, have well-founded foundations. The predictions of the model for the fluids' saturations do not differ substantially from what the Buckley–Leverett equation predicts, but the theory does not also need P_c and the RP curves as the input, which is a distinct advantage for the cases for which there are very little or no data. As such, Hilfer's theory is very appealing. Thus, further development of the theory is warranted.

14.16

Two-Phase Flow in Unconsolidated Porous Media

Two-phase flows through packed beds have been studied for decades as they are relevant to many phenomena of industrial importance. It is not possible to describe all aspects of the problem and the progress that has been made and, thus, we restrict our attention to some key points and concepts. There are many articles that review various aspects of two-phase flows through packed beds, including those of Satterfield (1975), Charpentier (1976), Herskowitz and Smith (1983), and the excellent paper of de Santos *et al.* (1991). More recent developments have been discussed by Toye *et al.* (1996) and Jiang *et al.* (2002).

In general, two-phase flows in packed beds operate on the principle of having sample-spanning fluid phases, despite having free surfaces between the two phases. As described by de Santos *et al.* (1991), the outcome of any two-phase flow phenomenon in a packed bed is the product of four factors.

1. The mass-transfer coefficient that measures the rate of transfer of a substance by convection and diffusion. Mass transfer depends on the interfacial area between the two phases – usually a liquid and a gas – and on the departure from equilibrium between them. Direct contact between the two phases can facilitate mass transfer, but this is not always useful because it can also lead to dispersing of one fluid in the other and formation of drops and bubbles, mists, and foams in the flowing fluids. Direct contact between the two phases is enhanced by local convection in the direction of diffusion. If this is to happen, one needs, near the phase boundary, an expandable and contractible interface, and an accelerating and decelerating surface velocity. This, in turn, requires varying cross

sections of the gas flow that, however, may lead to the local separation of the gas flow that makes the process inefficient.

2. The interfacial area per unit volume of the contacting particles.
3. The residence time of the fluids in the packed bed. For larger superficial velocities, the packed bed must be longer in order to achieve a certain residence time for a fluid phase that, in turn, requires a higher gas pressure drop and, thus, higher pumping cost.
4. The departure from mass-transfer equilibrium between the two fluid phases, which is measured in terms of the difference between the local concentrations in the two fluid phases. The concentration difference is often called the *driving force* in the chemical engineering literature. Perhaps the best way of maintaining the driving force at a certain level is by *countercurrent* flow of the two phases (see below). However, as pointed out by de Santos *et al.* (1991), this also causes a larger shear stress at the interface between the two fluids and, thus, a higher cost of pumping the fluids into the packed bed. In some cases, the driving force is maintained by a chemical reaction within a fluid phase, after a substance has been transferred from the other phase.

Similar to two-phase flows in pipes, there are several flow regimes in a packed bed that are described in detail by de Santos *et al.* (1991). A summary of their discussion is as follows.

14.16.1

Countercurrent Flows

If the flow rates of the two phases are low, then both fluid phases will be sample-spanning, also called *trickling*. If, however, the liquid-phase flow rate is larger than a certain critical value, then the amount of liquid that stays in the bed, and the pressure drop that is needed to drive the gas phase, both increase sharply. If the liquid-phase flow rate is high enough, the liquid accumulates at the top of the bed, a phenomenon that is called *flooding*. Under such conditions, the liquid can even be carried away by the gas phase, which can also happen if the liquid flow rate is fixed, and the gas flow rate is increased. Before the flooding occurs, the liquid begins to accumulate in a range called *loading*. Although loading might seem attractive because it increases the mass-transfer coefficient, it can also be counterproductive because it is close to the flooding that is not desirable, as flooding limits the flow rates at which the two phases can pass through the packed bed.

Various mechanisms have been proposed for flooding, with apparently no general agreement or consensus between them. For example, Shearer and Davidson (1965) proposed that a standing wave of flowing liquid is raised by the effect of counterflowing gas, and if the flow rate is high enough, the wave growth and the counterflowing gas reinforce each other to the extent that they block flow passages between the particles and induce flooding. On the other hand, Hutton *et al.* (1974) suggested that the onset of flooding is linked to the instability of liquid films that

flow down a solid wall, and the instability is accentuated by the shear stress induced by the counterflowing gas.

Factors that increase liquid holdup are both the liquid and gas flow rates, the liquid viscosity, and the porosity of the packed bed. Factors that decrease liquid holdup are liquid density, surface tension between the liquid and the gas, larger particles in the packing, and particles that are not wetted very well by the liquid. Moreover, as the liquid holdup increases, so also does the pressure difference needed to drive the gas. There have been many attempts to correlate the pressure difference with various factors, for example, the flow rates, fluid properties, and packing characteristics (see Ellman *et al.* (1988), for a review).

14.16.2

Cocurrent Downflows

There are at least four flow regimes associated with cocurrent downflows (Herskowitz and Smith, 1983) that are as follows.

1. *Trickling*, which happens when the liquid and gas flow rates are low enough. Then, the liquid flows as films over the solid particles, while the gas occupies and flows in the bulk of the pore space. An important consideration in this regime is the way the liquid is distributed in the system. If its flow rate is small enough, it can even flow through the localized rivulets, even if it does not completely wet the solid particles. Figure 14.12 presents the difference between film and rivulet flows. The latter is undesirable because it leads to poor contact between the fluids as well as between the fluids and the solid particles and, therefore, a smaller mass-transfer coefficient. It can also be caused by grooves, incomplete wetting, and capillary action.

Many groups (see, for example, Levec *et al.*, 1988) reported hysteresis loops in the trickling flow through a packed bed of particles with small diameters

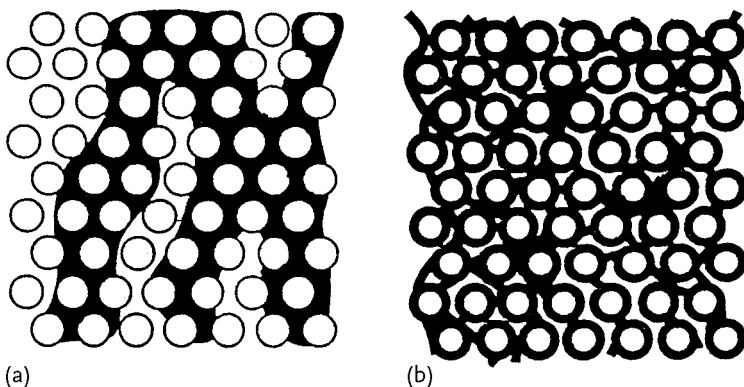


Figure 14.12 Rivulet (a) and film (b) flows in cocurrent downflows (after Christensen *et al.*, 1986).

(< 1.8 mm). More specifically, the pressure drop and liquid holdup for specified gas and liquid flow rates were found to depend on the manner by which they had been reached. This is, of course, similar to two-phase flows in consolidated porous media described earlier in this chapter. Figure 14.13 presents a typical hysteresis loop. The loop's upper branch corresponds to film flow in which all the particles are wetted by the liquid films. As described in Chapter 4 and earlier in the present chapter, the origin of such hysteresis loops is linked to the distribution of the liquid phase throughout the porous medium. Smaller particles enhance the influence of capillarity and, thus, the likelihood of hysteresis. Transverse dispersion can counter this effect.

2. *Pulsing*, which happens at intermediate values of the fluids flow rates. Under such conditions, liquid-rich and gas-rich slugs alternate as they travel down the bed, giving rise to pulsing discharge. The origin of pulsing flow and its characteristics have been studied by several research groups. Such a flow regime is characterized by large fluctuations in the pressure drop and liquid holdup. Several industrial reactors do, in fact, operate near the trickling-to-pulsing transition.
3. *Bubbling*, which occurs at high liquid and low gas flow rates. Under such conditions, the liquid controls the pore space and spans the system, while the gas phase is carried along in the form of small bubbles.
4. *Spray*, which happens at conditions opposite to those in bubbling. In this case, the gas phase spans the system, while the liquid phase is carried along in the form of dispersed droplets.

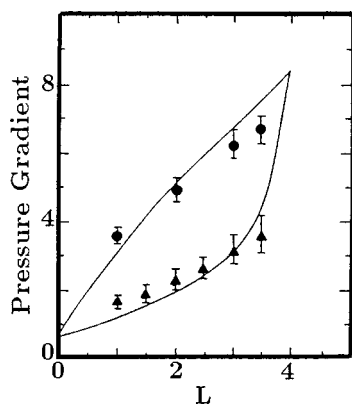


Figure 14.13 Comparison of the predicted pressure gradient (in kPa/m, shown with the symbols) with the experimental data. The gas flow rate is $756 \text{ kg/m}^2 \text{ h}$, and L is in $10^4 \text{ kg/m}^2 \text{ h}$ (after Chu and Ng, 1989).

Table 14.1 Values of the Reynolds number Re , the capillary number Ca , and the Bond number Bo that are typically encountered in two-phase flow through packed beds, and their comparison with those for consolidated porous media that are used in oil recovery processes.

Porous media	Re	Ca	Bo
Packed beds	10^{-2} – 10^3	10^{-1} – 10	10^{-1} – 10
Consolidated	10^{-9} – 10^{-2}	10^{-7} – 10^{-3}	10^{-9} – 10^{-2}

14.16.3

Cocurrent Upflows

In cocurrent upflows, both fluids enter the system from the bottom of the packed bed. Cocurrent upflows can be very useful operations if the ratio of gas to liquid flow rates is small, or if one needs a large residence time for the liquid. Turpin and Huntington (1967) identified three flow regimes for cocurrent upflows that are pulsing, bubbling, and spray, very similar to cocurrent downflows described earlier.

By now, it should be clear to the reader that two-phase flow of gases and liquids in packed beds involves a set of complex and fascinating phenomena. Moreover, as described earlier in this chapter, flow of two immiscible fluids in any porous medium involves competition between various forces and, as usual, the competition is expressed in terms of dimensionless groups. The relevant dimensionless groups are the Reynolds number Re , the capillary number Ca , and the Bond number Bo defined below by Eq. (14.46) below. Table 14.1, adopted from de Santos *et al.* (1991), presents the ranges of the three dimensionless groups for two-phase flow operations in packed beds, and compares them with those for consolidated porous media, typically encountered in oil recovery operations. As the table indicates, the ranges of the dimensionless groups that are used in the two types of porous media are vastly different, so much so that one cannot yield much insight into the other. The difference is one reason for separate studies of flow phenomena in unconsolidated porous media. It also indicates the rich variety of phenomena that one may observe in such porous media, which explains why flow in unconsolidated porous media has been of great interest for a long time.

14.17

Continuum Models of Two-Phase Flows in Unconsolidated Porous Media

As with all flow phenomena in porous media that we have studied so far in this book, two-phase flow in unconsolidated porous media has been modeled both by the continuum and pore network models. What follows is a brief description of the typical continuum models. Pore network models will be described in Chapter 15.

Continuum models of two-phase flow in packed beds are, to some extent, similar to those for two-phase flows in consolidated porous media. We describe a typical continuum model developed by Grosser *et al.* (1988) and Dankworth and Sundaresan (1989). There are many other such models in the literature, for which the interested reader can consult the two papers. The model that we describe consists of the volume-averaged equations of motion for each fluid phase (see Chapters 9–11), together with the appropriate constitutive equations. As discussed earlier in this chapter, the continuum models are valid if both fluid phases are sample-spanning and are not near macroscopic disconnection (the percolation threshold), and the linear size of the system (for example, the height of the packed bed) is much larger than the length scale over which the porous medium is homogeneous.

We assume that the volume fractions of the gas and liquid phases are ϕ_g and ϕ_l , respectively. Thus, we must have $\phi_g + \phi_l = \phi$, with ϕ being the porosity. The continuity and momentum equations for the fluid phases are given by

$$\frac{\partial \phi_i}{\partial t} + \nabla \cdot (\phi_i \mathbf{V}_i) = 0, \quad (14.41)$$

$$\rho_i \phi_i \left(\frac{\partial \mathbf{V}_i}{\partial t} + \mathbf{V}_i \cdot \nabla \mathbf{V}_i \right) = -\phi_i \nabla P_i + \phi_i \rho_i \mathbf{g} + \mathbf{F}_{di} + \phi_i \nabla \cdot \boldsymbol{\tau}_i + \nabla \cdot \mathbf{R}_i, \quad (14.42)$$

where i either denotes the liquid or the gas phase, $\mathbf{F}_{di}$ denotes the drag force exerted on phase i , $\boldsymbol{\tau}_i$ denotes the viscous stress tensor, and $\mathbf{R}_i$ denotes a pseudo-turbulence stress. Normally, $\nabla \cdot \boldsymbol{\tau}_i$ and $\mathbf{R}_i$ are small and can be safely ignored. The next step is to provide constitutive equations for the drag force $\mathbf{F}_{di}$. Many empirical or semi-empirical correlations have been proposed in the past that relate $\mathbf{F}_{di}$ to the flow velocity and the packed bed characteristics. In fact, the main difference between the various continuum models of two-phase flows in packed beds is mainly in the type of correlations that are used for $\mathbf{F}_{di}$. For example, Sáez and Carbonell (1985) proposed the following Ergun-type equations

$$\mathbf{F}_{dg} = \left[\frac{A\mu_g(1-\phi)^2\phi^{1.8}}{d_p^2\phi_g^{2.8}} + \frac{B\rho_g(1-\phi)\phi^{1.8}}{d_p\phi_g^{1.8}} |\mathbf{V}_g| \right] \mathbf{V}_g, \quad (14.43)$$

$$\mathbf{F}_{dl} = - \left(\frac{\phi - s_1}{\phi_l - s_1} \right)^{2.43} \left[\frac{A\mu_l(1-\phi)^2\phi_l^2}{d_p^2\phi^3} + \frac{B\rho_l(1-\phi)\phi_l^3}{d_p\phi^3} |\mathbf{V}_l| \right] \mathbf{V}_l. \quad (14.44)$$

Here, A and B are constants, and s_1 is called the *static holdup* and represents the volume fraction of the liquid in the packed bed in the absence of any flow. There are several empirical equations for s_1 . For example, it can be estimated from

$$s_1 = \frac{1}{20 + 0.9\text{Bo}}, \quad (14.45)$$

where Bo is the Bond number that, for a packed bed, is defined as

$$\text{Bo} = \frac{\rho_l g d_p^2 \phi^3}{\sigma_{lg}(1-\phi)^2}, \quad (14.46)$$

where σ_{lg} is the surface tension between the two fluids. Other correlations for the drag force were also given by Hutton and Leung (1974). The final step for formulating the continuum model is to provide a correlation, or else experimental data, for the capillary pressure P_c between the two phases. For example, Reed *et al.* (1987) measured P_c in beds of particles with up to 1 mm diameter, and found that the Leverett J -function (see Chapter 4) describes the capillary pressure well.

In practice, the above set of equations is too difficult to solve analytically or even numerically and, thus, one must make many simplifications. For example, it is typically assumed that the macroscopic flow is one-dimensional, and that the packed bed is long enough that the domain of the problem can be considered as infinite. As in any two-phase flow problem, one must also consider the possibility of instability phenomena (see Chapter 13 and the next section). Such continuum models predict that liquid holdup and pressure gradients below the flooding point can be approximated by a uniform state in which all temporal and spatial derivatives in the macroscopic equations vanish. They also predict the existence of two uniform solutions, a low holdup state that is commonly observed below the flooding point, and an upper holdup state that is not accessible to a steady flow. Thus, the maximum flooding conditions can be defined as those beyond which no uniform state solution exists.

Sáez and Carbonell (1985) and Levec *et al.* (1986) showed that Eqs. (14.43) and (14.44) provide satisfactory correlations for the pressure drop and liquid holdup in the cocurrent downflow regime, and Dimenstein and Ng (1986) and Sundaresan (1987) were able to describe the size, velocity, and holdup of pulses in cocurrent downflows by using such continuum models. Thus, for flow in packed beds, the continuum models have been relatively successful. One reason for the success is that in both the rivulet and film flows, one deals with a percolation phenomenon far from the percolation threshold and, thus, the continuum models are expected to be relatively accurate.

14.18

Stability Analysis of Immiscible Displacements

In this section, we carry out an analysis of the stability of immiscible displacements in porous media. However, we first provide a qualitative discussion of the subject, and then follow it up with a more quantitative analysis. Over 50 years ago, van Meurs (1957) used displacing and displaced fluids of the same refraction index and studied immiscible displacements in porous media. Chouke *et al.* (1959), Perkins and Johnston (1969), White *et al.* (1976), Peters and Flock (1981), Paterson *et al.* (1982, 1984a,b), Måløy *et al.* (1985, 1987), Stokes *et al.* (1986), Frette *et al.* (1990), and Oxaal (1991) provided more experimental results and insight. The data and the observations indicate that immiscible fingering can occur over many length scales, up to a macroscopic one and, therefore, one may even use a characteristic length scale for characterizing what is seen in experiments. The main implications of most of the experimental works are as follows.

1. If the invading fluid wets the porous medium, fingering is characterized by some macroscopic length scale, such as, the width of the fingers, whereas if it does not, fingering is limited to pore scales in which case shielding dominates spreading (see Chapter 13). Recall that a displacement with a wetting fluid is compact. The characteristic macroscopic length scale decreases as the capillary number Ca increases. Careful experiments of Stokes *et al.* (1986) confirmed these observation. Stokes *et al.* (1986) also showed that when macroscopic fingering occurs, the width w of the finger scales with the permeability and Ca as $w/\sqrt{K_e} \sim 1/\sqrt{Ca}$.
2. If the invading fluid is non-wetting, then one obtains percolating fingers. This phenomenon will be studied in detail in Chapter 15.
3. There is a transition zone just behind the interface where both fluid phases are flowing.

Chouke *et al.* (1959) were the first to attempt a theoretical analysis of stability of two-phase flows in a porous medium. They ignored the transition zone and assumed that one fluid completely displaces another fluid. As the boundary condition at the interface, they used $P(r) = a_1\kappa + a_2$, but replaced the microscopic surface tension by an *effective macroscopic* surface tension. Although there is no theoretical justification for doing this (as the surface tension operates at the pore scale), the macroscopic surface tension did provide a reasonable description of some of the experimental data. Others (Outmans, 1962; Rachford, 1964; Hagoort, 1974; Peters and Flock, 1981; Huang *et al.*, 1984; Jerauld *et al.*, 1984a,c; King *et al.*, 1984; Yortsos and Huang, 1986; Chikhliwala and Yortsos, 1988; Chikhliwala *et al.*, 1988; Yortsos and Hickernell, 1989; Riaz and Tchelepi, 2004) analyzed stability of immiscible displacements in porous media.

We consider the results that are obtained from displacements at a constant velocity V of a non-wetting fluid by the injection of a wetting fluid. The system is initially at a uniform saturation S_{iw} . Unidirectional flow is described by the equation

$$\phi \frac{\partial S_w}{\partial t} + V \frac{\partial f_w}{\partial S_w} \cdot \frac{\partial S_w}{\partial z} = - \frac{\partial}{\partial z} \left(f_w \frac{K_e k_{rw}}{\mu_w} \frac{dP_c}{dS_w} \cdot \frac{\partial S_w}{\partial z} \right), \quad (14.47)$$

with the initial and boundary conditions, $S_w = S_{iw}$, $t = 0$, and

$$f_w \left(1 + \frac{K_e k_{rw}}{V \mu_{nw}} \cdot \frac{dP_c}{dS_w} \frac{\partial S_w}{\partial z} \right) = f_0 \quad z = 0, \quad (14.48)$$

where f_w is the fractional flow of the wetting phase. If f_w has an upward convex segment, which is usually the case during imbibition or during drainage of not strongly wetting fluids, then the base state is taken as the steady-state solution of Eq. (14.47).

Various upstream conditions, denoted by $-\infty$, are graphically shown in Figure 14.14. We use dimensionless notations to simplify the discussion; that is, a reduced saturation

$$S_{rd} = \frac{S_w - S_w^\infty}{S_w^{-\infty} - S_w^\infty}, \quad (14.49)$$

and normalized mobilities (see Chapter 13),

$$\lambda_w(S_{rd}) = \frac{k_{rw}}{k_{rw}^{-\infty}}, \quad \lambda_{nw}(S_{rd}) = \frac{k_{rnw}}{k_{rnw}^{-\infty}}, \quad (14.50)$$

$$M = \frac{k_{rw}^{-\infty}}{k_{rnw}^{-\infty}} \frac{\mu_{nw}}{\mu_w}, \quad \lambda_t = M\lambda_w + \lambda_{nw}, \quad (14.51)$$

$$\lambda_c = \sqrt{\frac{K_e}{\phi}} \frac{1}{\sigma_{wnw} \cos \theta} \frac{dP_c}{dS_w} < 0. \quad (14.52)$$

All the length scales are measured by $L = \sqrt{K_e/\phi} (S_w^{-\infty} - S_w^\infty) k_{rw}^{-\infty} / Ca$, which is the length over which the viscous and capillary forces are balanced. In the new notation, we have

$$\lambda_{nw} \lambda_w \lambda_c \frac{dS_{rd}}{d\xi} = \lambda_t (f S_{rd} + f_w^\infty) - M \lambda_w, \quad (14.53)$$

$$\lambda_{nw} \frac{dP_c}{d\xi} = f S_{rd} + f_w^\infty - 1, \quad (14.54)$$

where $f = f_w^{-\infty} - f_w^\infty$, and $\xi = z/L$. We note that if $k_{rw}(S_w = S_{iw}) > 0$, that is, if the saturation is mobile, then the downstream decay of the base state is exponential. In the opposite case, we have

$$\frac{dS_{rd}}{d\xi} \sim -S_{rd}^{-m}, \quad (14.55)$$

where $0 < m < 1$ for the secondary displacements, and $m > 1$ for the primary displacements. Equation (14.55) is a manifestation of hypodiffusion described earlier. To examine the stability of the displacement, Eqs. (14.53) and (14.54) are written in terms of a moving coordinate system, and the stability of the resulting equations is analyzed using the general method described in Chapter 13. The following results are obtained.

1. The rate of growth ω is bounded from above for all wave numbers α by the expression

$$\omega < -A_1 \alpha^2 + A_2 A_3 \alpha, \quad (14.56)$$

where $A_1 = \min(-\lambda_{nw} \lambda_w \lambda_c / \lambda_t) > 0$, $A_2 = \max(df_w/dS_{rd}) > 0$, and $A_3 = \lambda_t(1)/\lambda_t(0) - 1 > 0$, where one and zero correspond to upstream and downstream values of S_{rd} , respectively (see Eq. (14.49)). Therefore, ω lies below the parabola on the right side of Eq. (14.56) and a cutoff wave number $\alpha_{co} \leq A_2 A_3 / A_1$, whereas the maximum growth rate ω_m does not exceed $(A_1 A_3)^2 / (4A_1)$. These general results confirm what is expected, namely, that long-wave instability is driven by mobility contrast (A_3), whereas short-wave stabilization is a result of capillarity (A_1).

2. One can develop large wavelength asymptotics, $\omega = \omega_1 \alpha + \omega_2 \alpha^2 + \dots$, where

$$\omega_1 = f \frac{\lambda_t(1) - \lambda_t(0)}{\lambda_t(1) + \lambda_t(0)}, \quad (14.57)$$

which is a generalization of the Saffman–Taylor condition, $\omega = \alpha(M-1)/(M+1)$, for the Hele–Shaw cells mentioned in Chapter 13. ω_2 is not zero, but varies according to the upstream decay of the base state. For example, if the upstream decay is algebraic (see Figure 14.14a), then one has $\omega = \omega_1 \alpha + \omega_2^* \alpha^2 \ln \alpha + \omega_3 \alpha^2 + \dots$, where

$$\omega_2^* = \frac{4 \left[\lambda_t(0) \frac{d\lambda_t}{dS_d} \right] |_{S_d=1}}{k [\lambda_t(0) + \lambda_t(1)]}, \quad (14.58)$$

where k is a measure of the algebraic decay. Since $[d\lambda_t/dS_d]|_{S_d=1} > 0$ for unstable displacements, its existence indicates a stabilizing effect. Interestingly, this is due to the mobility effect alone that adds to the process stability. Chikhliwala and Yortsos (1988) obtained the numerical solutions of Eqs. (14.53) and (14.54) in order to demonstrate the adequacy of the expansions.

The geometries of the interface and the medium can have an important effect on the stability. For miscible displacements, Tan and Homsy (1987) suggested an algebraic (rather than exponential) dependence for the time evolution of the disturbances, which was then applied by Yortsos (1987b) to immiscible fluids in radial displacements. In this case, the base-state profiles are sole functions of the similar-

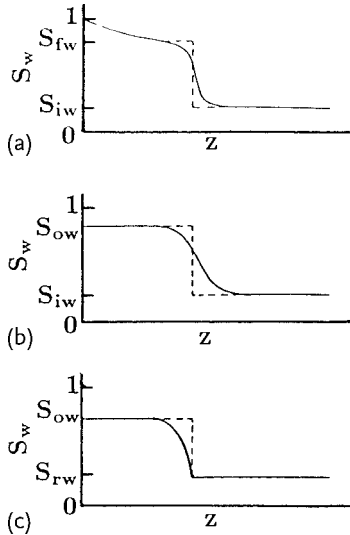


Figure 14.14 Various upstream conditions for the wetting-phase saturation S_w , versus the axial distance z , used in the stability analysis of immiscible flows.

ity variable, $\xi_s = r^2/t$. Perturbations in the following forms are then sought,

$$S_{rd} = \bar{S}(\xi_s) + t^\delta s(\xi_s) \exp(i\alpha\chi), \quad (14.59)$$

$$P = \bar{P}(\xi_s) + t^\delta p(\xi_s) \exp(i\alpha\chi), \quad (14.60)$$

where $\bar{P}$ and P are dimensionless capillary pressures, χ is the azimuthal angle, and the bars indicate the mean (unperturbed) values (see Chapter 13). If $\delta > 0$, then the interface is unstable. For small α , one has the asymptotic expansion

$$\delta = -1 + \delta_1\alpha + \delta_2\alpha^2 + \dots, \quad (14.61)$$

where δ_1 is equivalent to the Saffman–Taylor term, that is, $\omega = \alpha(M-1)/(M+1)$, and δ_2 is inversely proportional to a capillary number Ca_m . Large Ca_m leads to instability, but if Ca_m is not too large, even $M > 1$ may not lead to instability. For $Ca_m < Ca_c$, the displacement is *stable*, where Ca_c is given by $Ca_c = 2f_w/\omega_m$, and ω_m is a parameter independent of Ca . It should be pointed out that as Yortsos (1987a) showed, immiscible displacements are equivalent to miscible displacements *with equilibrium adsorption*. Such an analogy relates (S, f_w) to the flowing and adsorbed concentrations (C_f, C_a) .

What was presented so far represents a linear stability analysis. Clearly, accurate numerical simulations must be carried out in order to not only confirm the predictions of the linear stability analysis, but also understand the nonlinear regime that develops at longer times. Such high-resolution numerical simulations of immiscible two-phase flow in porous media were carried out by Riaz and Tchelepi (2006a), which confirmed the predictions of the linear stability analysis for short times. In addition, the simulations indicated that the fingers interacted much more weakly than those in miscible displacements. The wave number of the nonlinear fingers was found to rapidly decrease due to shielding (see Chapter 13), and scale uniformly as t^{-1} at long times. The dominant feature for the flows was found to be the gravity tongue.

Riaz *et al.* (2007) presented a set of accurate experimental data and visualization, coupled with linear stability analysis and high-resolution numerical simulations. Their work clearly demonstrated that the decline in oil production at larger capillary numbers is linked with viscous instability. Moreover, the Buckley–Leverett model described earlier was found to provide an accurate description of the displacement only when the flow is stable, but that it also accurately captures oil production at long times *after* the breakthrough when the unstable front has moved out of the porous medium.

The analysis presented can be extended in several directions. Riaz and Tchelepi (2004) extended the linear stability analysis to the case in which both capillary dispersion and density variations exist. Density variations is also important to a variety of displacement problems, such as, sequestration of carbon dioxide in saline aquifers (Riaz *et al.*, 2006). Riaz and Tchelepi (2006a) showed through high-resolution numerical simulations that even a small density variation can eliminate viscous fingers.

As the discussions of the properties of the relative permeabilities indicated, the RP curves can take on a wide variety of shapes. Therefore, it is also important to study the effect of the RP curves on the stability characteristics of immiscible flows. Riaz and Tchelepi (2006b) carried out such a study by using the RP curves that were functionally similar to Eqs. (14.22) and (14.25). By changing the exponents in Eqs. (14.22) and (14.25), one obtains a variety of the RP curves. Riaz and Tchelepi (2006b) carried out a linear stability analysis and showed that viscous instability of two-phase immiscible fluids are governed not only by the end point values of the RPs, but also by their actual profiles. Moreover, the RP functions that are associated with drainage yield a more unstable displacement than the functions that are attributed to imbibition. This would not be surprising (although not noted prior to the analysis of Riaz and Tchelepi) if we recall that imbibition results in compact clusters of fluids, whereas drainage gives rise to fractal-like structures and, as discussed in Chapter 13 (see also Chapter 15 for a comprehensive discussion), fractal structures are usually associated with unstable flows. High-resolution numerical simulations of Riaz and Tchelepi (2006b), well into the nonlinear regime, indicated that the finger amplitude initially grows at a rate proportional to $t^{1/2}$, then drops to $t^{1/4}$ at later times, and finally grows linearly with t .

14.19

Two-Phase Flow in Large-Scale Porous Media

So far, the discussion has been restricted to laboratory-scale porous media. However, similar to every phenomenon studied so far in this book, two-phase flows in large-scale (LS) porous media have also been studied both experimentally and by numerical simulations. Let us first describe a few key experimental results, and then describe the theoretical studies.

An important characteristic of any LS porous medium is its stratification. Almost all the LS natural porous media contain layers of contrasting properties that not only make the media globally anisotropic, but also add much complexity to any theoretical or computer simulation studies of fluid flow in such porous media. Thus, one way of studying two-phase flow in the LS porous media is by experimenting with a porous medium – even if at a small laboratory scale, which contains some contrasting layers.

Ogandzanjanc (1960) was perhaps the first to experimentally study flow in a stratified porous medium, experimenting with an unconsolidated porous medium with two layers. His experiments indicated that there is significant *crossflow* between the strata. Moreover, initially, the flow velocity was higher in the more permeable layer (as is expected), and that flow in each layer was similar to a single-phase flow system. Due to the crossflow, however, the distance between the interfaces in the two layers stabilized. Ever since Ogandzanjanc's work, there have been several other experimental studies, including those of Novosad *et al.* (1984), Sorbie *et al.* (1987), Ahmed *et al.* (1988), and Bertin *et al.* (1990). The last group studied waterflooding in a porous medium with two strata, with one stratum made of Aerolith-10, an ar-

tificial sintered porous medium, and with the second one being Berea sandstone. The two strata had the same thickness. The results indicated the strong effect of heterogeneities within each stratum and the contrast between them on the performance of the waterflood and the volume fraction of the recovered oil. Bertin *et al.* (1990) also found that even small scale heterogeneities within the sandstone layer could strongly affect the waterflood process.

Most of the theoretical studies of two-phase flow in stratified porous media have two goals. One is to examine the crossflow between the strata, its significance, and its variations with the time as the displacement proceeds. The second goal is to determine the important properties of the displacement process. For example, Douglas *et al.* (1959) studied imbibition in a layered porous medium and used an averaged form of Darcy's law to describe the two-phase flow problem. Goddin *et al.* (1966) studied waterflood processes in stratified media and concluded that crossflows can be caused by both capillary and viscous forces, while Yokoyama and Lake (1981) undertook an extensive study of the effect of capillary forces on crossflows.

Kyte and Berry (1975) used large-scale numerical simulation to study immiscible displacements in stratified media, while Coats *et al.* (1971) used a hydrostatic distribution of various fluid phases in the vertical direction to obtain large-scale capillary pressure and relative permeability curves for the stratified medium that they utilized.

14.19.1

Large-Scale Averaging

Perhaps the simplest heterogeneous system to consider is a stratified porous medium with several layers, but with no crossflow between the layers. Each stratum is characterized by an effective permeability. Dykstra and Parsons (1950) considered two-phase flow in such a porous medium and, assuming that flow in each stratum was piston-like (constant velocity), derived an expression for the amount of recovered oil just at the breakthrough point, that is, at the point where the interface in one of the strata reaches the outlet of the system. Reznik *et al.* (1984) generalized the Dykstra–Parson model and derived expressions that can be used for calculating the amount of oil recovered at any stage of the process, and the corresponding *large-scale* RPs at that stage. If we do calculate the RPs, we find that the shape of the resulting RP curves are *not* similar to those obtained with laboratory-scale porous media that were described earlier in this chapter.

The literature on two-phase flow in stratified media with communicating layers is relatively extensive. Starting with Goddin *et al.* (1966), many groups (Coats *et al.*, 1971; Martin, 1968; Hearn, 1971; Jacks *et al.*, 1973; Kyte and Berry, 1975; Killough and Foster, 1979; Yokoyama and Lake, 1981; Kortekaas, 1983; Wright and Dawe, 1983; Ypma, 1983; Bertin *et al.*, 1990) studied two-phase flows in stratified porous media using numerical simulations. To give the reader some idea as to the number of simulations carried out, we consider a 2D porous medium with only two layers. Suppose that the two fluids are water and oil, and that they and the porous medium

are incompressible. Writing a material balance for the water phase, we obtain

$$\frac{\partial}{\partial x} \left[\frac{K_{xw}}{\mu_w} \left(\frac{\partial P_0}{\partial x} - \frac{\partial P_c}{\partial x} \right) \right] + \frac{\partial}{\partial y} \left[\frac{K_{yw}}{\mu_w} \left(\frac{\partial P_0}{\partial y} - \frac{\partial P_c}{\partial y} \right) \right] = \phi \frac{\partial S_w}{\partial t}, \quad (14.62)$$

where P_0 is the pressure in the oil phase, and K_{xw} and K_{yw} are the water phase permeabilities in the x - and y -directions (recall that a stratified porous medium is anisotropic). If the functional forms for K_{xw} , K_{yw} and P_c are assumed, then Eq. (14.62) can be solved numerically for various boundary conditions at the interface between the two layers, and the injection conditions.

If the layers are homogeneous, then the results of the previous sections can be used for K_{xw} , K_{yw} and P_c within each layer. The crossflows between the layers can be of various natures. One may have systems with only viscous crossflows, or with viscous and capillary crossflows, and so on. If we rewrite Eq. (14.62) in a dimensionless form, then *direction-dependent* capillary numbers emerge. The two capillary numbers Ca_x and Ca_y would be related to one another by

$$Ca_y = Ca_x R_L^2, \quad R_L^2 = \left(\frac{L}{H} \right)^2 \left(\frac{K_y^0}{K_x^0} \right)^{1/2}, \quad (14.63)$$

where L and H are the length and thickness of the medium, and K_x^0 and K_y^0 are the permeabilities of a reference phase (which can be taken to be either oil or water).

Normally, the results of the numerical simulations are averaged over the vertical direction. If this is done, then one ends up with quantities that are referred to as *pseudo functions* in the petroleum engineering literature. For example, one can use pseudo functions for the RPs and another pseudo function for the capillary pressure. Such functions are, in fact, nothing but what Quintard and Whitaker (1988) refer to as *large-scale* RPs and capillary pressure, that is, the RPs and capillary pressure for a LS porous medium that, in general, may or may not be anisotropic or stratified. From a scientific point of view, *large-scale functions* are much more appealing than pseudo functions since, as Quintard and Whitaker (1988) pointed out, the word *pseudo* suggests that the functions are something less than what they purport to be, whereas the analysis of Quintard and Whitaker (see below) indicates that such functions can be deduced from a rigorous analysis for *any* LS porous medium.

Quintard and Whitaker (1988) developed a LS averaging technique for two-phase flow in megascopic porous media. Starting with Eqs. (14.17)–(14.20) as the *locally-averaged* equations, they developed LS averaged continuity and momentum equations for each fluid phase, allowing for the possibility that a portion of a fluid phase may be trapped by another fluid phase, which happens when an incompressible fluid is surrounded by another immiscible fluid. In order to make the theory tractable, Quintard and Whitaker assumed that the system is in local mechanical equilibrium, implying that the *local* fluid distribution is determined by capillary pressure–saturation relations, and is not limited by the solution of an evolutionary transport equation.

In two subsequent papers, Quintard and Whitaker studied the effect of the LS spatial and temporal gradients (Quintard and Whitaker, 1990a), and investigated two-phase flow in a heterogeneous and stratified medium under the quasi-static and dynamic conditions (Quintard and Whitaker, 1990b). The quasi-static condition in the present context means that, the local capillary pressure, everywhere in the averaging volume, is equal to the LS P_c evaluated at the centroid of the averaging volume, and that the LS P_c is given by the difference between the LS pressures in the two fluid phases. As such, the LS P_c is assumed to be independent of such complex factors as transient, gravitational and flow effects. If there is significant departure from the quasi-static conditions, then one has a displacement. Quintard and Whitaker found that even at relatively low flow rates, dynamic effects may be important. Bertin *et al.* (1990) compared the predictions of the theory of Quintard and Whitaker with their experimental data, but only found qualitative agreement. Ahmadi and Quintard (1993) made a comprehensive comparison between the predictions of the LS averaged equations and those obtained by the pseudo functions and other upscaling methods.

14.19.2

Reservoir Simulation

The petroleum engineering literature is, of course, replete with hundreds, if not thousands, of papers that describe various approaches to what is popularly called *reservoir simulations*, which are nothing but numerical simulations of two- or three-phase flows in oil reservoirs that represent LS heterogeneous porous media. Reservoir simulators that are typically used in the petroleum industry are usually one of two types. The first type is the so-called *black oil* simulators in which it is assumed that the fluids (usually oil, gas, and water) are homogeneous, and that the gas can dissolve in the oil, or vice versa, in *any* proportion. Such an assumption avoids the problem of computing the detailed phase diagrams of the mixture that requires an accurate thermodynamic equation of state. The second type of simulator is more complex; they are usually called *compositional simulators*. Such simulators represent the oil as a mixture of a few hydrocarbons and perform detailed phase equilibria calculations in order to determine the distribution of the fluids in the liquid and vapor phases in the reservoir. Both simulators use a huge amount of computer time, and for this reason, devising efficient numerical methods for solving the transport equations has always been an active area of research.

We do not intend to describe all the numerical methods that have been proposed for reservoir simulation since the subject deserves to have a separate *book* by itself. There are already many good books on the subject (see, for example, Fanchi, 2006). Several reviews also discuss the subject, such as Christie (1988) and Shiles *et al.* (1990) and, more recently, that of Sahimi *et al.* (2010).

Two issues are of prime importance to efficient reservoir simulations. One is the structure of the computational grid and the numerical methods that are used for solving the governing equations. Simulation of multiphase flows in porous media involves solving a parabolic pressure equation and a hyperbolic system of conser-

vation laws. Most reservoir simulators are based on a finite-difference (FD) approximation of the governing equations. The reservoir is divided into blocks, and the flow field is computed between the blocks. In the simulations of multiphase flows in the LS porous media, for example, oil reservoirs, the heterogeneities give rise to large gradients in the fluids' velocities, saturations, and the capillary pressure, especially around wellbores or in the vicinity of fractures. Given that, from a practical point of view, these are in fact the most interesting areas, the standard (five- or seven-point) FD approximation can result in unphysical solutions. Moreover, the solution may not be accurate enough, especially if large blocks are used. One may also have *numerical dispersion* (Lantz, 1971), which can mimic *physical dispersion*. While development of numerical dispersion can be advantageous, if physical dispersion is actually present, it can also be a disadvantage in the sense of producing solutions that do not actually mimic the true situation. They may also lead to spurious oscillations or overshoots in the neighborhood of high-gradient areas that result in unstable solutions and unphysical results. A variety of methods have been suggested for addressing such problems, including using finer grids where high gradients develop, unstructured grids, and so on (see Hoteit and Firoozabadi, 2008, and references therein).

The second issue of importance in reservoir simulations is the so-called *upscaling problem*, which is as follows. Given that the data for the RPs and capillary pressures are obtained with small, laboratory-scale porous media, how should one upscale such data from the small scale to the much larger scales that can be used in oil reservoir simulations? In the computational grids used in reservoir simulations, the grid blocks' size may be up to tens of meters, whereas the cores with which the data are measured are much smaller, by at least one order of magnitude or more. The LS averaging, the pseudo-functions, and more recent methods, such as those based on multiresolution wavelet transformations (Mehrabi and Sahimi, 1997; Ebrahimi and Sahimi, 2002, 2004; Rasaei and Sahimi, 2008, 2009a,b; Sahimi *et al.*, 2010), all represents the attempts to address the issue.

14.20

Two-Phase Flow in Fractured Porous Media

Two-phase flow in fractured rock is important to enhanced oil and gas recovery, isolation of radioactive waste, exploitation of geothermal fields for producing electricity, and recovery of coalbed methane. Compared with our knowledge about two-phase flow in unfractured porous media, not much is known about the same phenomena in fractured ones. Multiphase flow within a fracture and the exchange between the fracture and the porous matrix are controlled by the structure of the porous medium and its fracture network, the flow rate, and capillary and viscous forces. If the porous medium is water-wet, capillary pressure drives water from the fractures to the matrix, whereas viscous forces favor flow through the fractures.

If we inject water into a porous sample that contains a single fracture and is saturated with oil, then, depending on the flow rate, we may have high or low oil

recovery. At low flow rates – or low capillary number Ca – the water flows preferentially through the matrix, hence resulting in high recoveries. At higher flow rates or capillary numbers, the water will percolate through the fracture, leaving behind a large amount of oil. Thus, the recovery is a strong function of the capillary number.

For a typical oil reservoir, the capillary number Ca is about 10^{-7} or lower. However, because flow velocities in laboratory-scale porous media are larger than those in the reservoir, the typical Ca for such media is 10^{-6} – 10^{-4} . For oil and gas systems, the capillary number is about 10^{-5} – 10^{-2} due to the low surface tension between the two fluids. On the other hand, compared with porous matrices, the flow rates are high in fractures, hence giving rise to capillary numbers that are around 10^{-5} away from the wells, and 10^{-3} near wells. As pointed out earlier, once the capillary number exceeds 10^{-5} , viscous forces become important and cannot be ignored.

Despite its severe shortcomings that were described in Chapter 12, in the oil industry, the standard model for simulating two-phase flows in fractured porous media is the double-porosity or the double-permeability model. The appropriate governing equations for two-phase flows in the double-porosity model are given by Kazemi and co-workers (Kazemi *et al.*, 1969, 1976; Kazemi and Gilman, 1993) and, therefore, will not be given here. However, the model is oversimplified, particularly if, as described in Chapters 6, 8, and 12, the porous medium contains a network of interconnected fractures. In addition, the method is essentially a lumped-parameter approach in which the saturations, flow, and history-dependence of the exchange between the matrix and fractures all are represented by the RPs k_r , unless k_r and other parameters are adjusted. While the adjusted parameters may be able to provide reasonable fit of the existing data, they usually lack any predictive ability. Also, recall that two-phase flows in porous media crucially depend on the connectivity of the fluid phases, and that double-porosity models completely ignore this important effect.

The key to the double-porosity model is the so-called *transfer function* that expresses the exchange between the matrix and fractures (see Chapter 12). In order to improve the predictive power of the double-porosity model, many attempts have been made to develop more realistic expressions for the transfer functions (see, for example, Dutra and Aziz, 1992).

Even if the lumped-parameter approach with the RPs k_r could work, the fact is that our current understanding of the RPs for fractures is far from complete. Typically, straight-line RP curves are used, partly based on the experimental work of Romm (1966) in which oil and water were confined to different regions of a *smooth* fracture by controlling the wettability of the fracture surface. However, the experimental work of Merrill (1975) and theoretical analysis and numerical simulations of Pruess and Tsang (1990) for *rough* fractures indicated that the assumption of linear RPs may be in serious error, and that Romm's results were specific to his experimental system and have no general validity; see also Akin (2001).

15

Immiscible Displacements and Multiphase Flows: Network Models

Introduction

In Chapter 14, we described the essential physics of multiphase flows in disordered porous media. As discussed there, a large number of factors affect this class of phenomena, including capillary, viscous, and gravitational forces, the viscosities of the fluids and the interfacial tension separating them, the properties of the pores' or fractures' surface, the shape, size and connectivity of the pores (or fractures), and the wettability of the fluids. In Chapter 14, we also described the continuum models of multiphase flows in porous media.

As described in Chapter 3, whenever we deal with a disordered multiphase system, the connectivity of its phases plays a crucial role in determining its macroscopic properties, and multiphase flows in porous media are no exception. The continuum models that were described in Chapter 14 have provided a wealth of information and insight into the phenomena of multiphase flows in porous media, but they cannot predict the relative permeabilities (RPs) to the flowing phases (see, however, Hilfer, 2006a,b,c) which crucially depend on the connectivity of the pore space, the pores' shape, and the wettability of the fluids. In fact, the RPs and capillary pressure represent the input to such models. Therefore, one must develop an independent way of computing the RPs and the capillary pressure. Pore network models, of the type described in Chapters 7, 10, and 11, currently represent the most promising approach to the task of computing the RPs and the capillary pressure. Modeling and computing the latter was already described in Chapter 4. In the present chapter, we describe the pore network models of multiphase flows and the computations of the RPs.

15.1

Pore Network Models of Capillary-Controlled Two-Phase Flow

Some of the pore network models are explicitly based on the percolation concepts and their variants and, strictly speaking, are applicable only when the capillary number Ca is very small. Other models, though more general and applicable even when the capillary number is not too small, are still based on the percola-

tion concepts as they still invoke the concept of macroscopic connectivity. The limit $Ca \rightarrow \infty$ represents, of course, miscible displacements already described and studied in Chapter 13.

15.1.1

Random-Percolation Models

We should point out at the outset that a fundamental assumption in *all* the percolation models of two-phase flow in porous media is that the bond or site occupation probability p , that is, the probability that a bond or site is filled by a fluid, is proportional to the capillary pressure needed for entering that bond or site. Without such an assumption, it would be difficult to make a one-to-one correspondence between a percolation model and the two-phase flow problem. Although in some percolation models, such as, invasion percolation (see below), the occupation probability is not explicitly defined, its analog can be readily calculated.

The first random percolation model of two-phase flows in porous media was suggested by Larson (1977), with the details given in a series of papers by Larson *et al.* (1977, 1981a,b). Larson *et al.* (1981a) proposed a model for drainage, that is, displacement of a wetting fluid by a non-wetting one. The porous medium was represented as a simple-cubic network of bonds and sites with distributed sizes. It was assumed that a bond next to the interface is penetrated by the displacing fluid if the capillary pressure at that point exceeds a critical value, implying that the bond's radius must exceed a critical radius $r_{\min}$, the same radius that is defined by Eq. (4.52), which implies that during drainage, the *largest* pore throats are invaded by the non-wetting fluid. All the bonds that are connected to the (non-wetting) displacing fluid by a path of pores or bonds with effective radii larger than $r_{\min}$ are considered as *accessible*, with the accessibility being defined in the percolation sense described in Chapter 3. It was also assumed that all the accessible bonds with radii that are at least as large as $r_{\min}$ are filled with the non-wetting fluid. The assumption is not, of course, correct, as an interface that starts at one external face of a porous medium must travel along a certain path before it reaches an accessible bond that can be penetrated. Larson *et al.* (1981a) also assumed that the displaced fluid is compressible, so that even if a cluster of pore throats filled by the fluid is surrounded by the displacing fluid, it can still be invaded. As we discuss below, the assumption of compressibility does not, however, result in a serious error.

Larson *et al.* (1981b) proposed a percolation model of imbibition in order to calculate the residual non-wetting phase saturation S_{rnw} and its dependence on the Ca . To do so, they modeled the creation of isolated blobs of the non-wetting fluid by a random site percolation (see Chapter 3), and calculated the fraction $\hat{g}(s)$ of the active sites at the site percolation threshold that are in clusters of length s in the direction of flow. Larson *et al.* argued that the quantity represents the desired blob size distribution. To compute S_{rnw} , they assumed that once a blob is mobilized, it is permanently displaced. However, as discussed in Chapter 14, this is not always the case because a blob can get trapped again, can join another blob to create a larger one, and so on.

The fundamental assumption in the work of Larson *et al.* is that pore-level events are controlled by the capillary forces. Let us employ simple scaling arguments to estimate the values of the capillary number for which the fundamental assumption is valid. The capillary pressure across the interface is proportional to

$$P_c \sim \frac{\sigma \cos \theta}{\ell_g}, \quad (15.1)$$

where ℓ_g is a typical grain size, σ is the interfacial tension, and θ is the contact angle. On the other hand, the viscous pressure drop is proportional to

$$P_\mu \sim \frac{\mu_w v \ell_g}{K_e}. \quad (15.2)$$

Therefore,

$$\frac{P_\mu}{P_c} \sim \frac{Ca}{K_d}, \quad (15.3)$$

where $K_d = K_e/\ell_g^2$ is a dimensionless permeability which is small (on the order of 10^{-3} or smaller) because K_e , the effective permeability of the porous medium, is controlled by the narrowest throats in the medium. It follows that for capillary-controlled displacements, one must have $Ca \ll 1$. In practice, one has $Ca \sim 10^{-6} - 10^{-8}$. Experimental data (Le Febvre du Prey, 1973; Amaefule and Handy, 1982; Chatzis and Morrow, 1984) seem to support the estimate since, as discussed in Chapter 14 (see Figure 14.7), they indicate that S_{rnw} is constant for $Ca < Ca_c$, where Ca_c is the critical value of Ca for capillary-controlled displacement, whereas S_{rnw} only decreases when $Ca > Ca_c$. Larson *et al.* (1981b) compiled a wide variety of experimental data and compared them with their predictions.

Heiba *et al.* (1982, 1983, 1984, 1992) further developed the random percolation model and used it to compute the RPs for all regimes of the wettability described in Chapter 14. They distinguished between pore throats that are *allowed* to a fluid – that is, can potentially be filled by the fluid – and those that are actually occupied by it. Then, given the pore size distribution (PSD) of the pore space, they derived the PSD of the allowed and occupied pores.

Consider, for example, a displacement process in which one fluid is strongly wetting, while the other one is completely non-wetting. Then, according to the percolation model of Heiba *et al.* (1982, 1992)¹⁾, during the primary drainage, the PSD of the pores occupied by the displacing (non-wetting) fluid is given by Eq. (4.53), since the *largest* throats are occupied by the non-wetting fluid, whereas during imbibition, the PSD of the pores occupied by the displacing (wetting) fluid is given by Eq. (4.56), because the *smallest* pore bodies are occupied by the wetting fluid. One can, in a similar fashion, derive expressions for the PSD of the pores occupied by

1) The original work was completed in early 1982, with the results presented as a preprint at a 1982 SPE conference, and accepted for publication in the same year, subject to some minor clarifications. It took, however, ten years to make the clarification since the

first two authors had moved on, and the last two were preoccupied with other things! In the meantime, a whole “industry” had been created based on the preprint! The paper was eventually published in 1992, setting a world record for delay!

the displacing and displaced fluids during the secondary imbibition and drainage. Once the PSDs are determined, calculating the permeability of each fluid phase and, therefore, its RP, reduces to a problem of percolation conductivity because when the permeability of a given fluid phase is computed, the conductance (or effective radii) of the bonds occupied by the second fluid can be set to zero as the two phases are immiscible. This assumption neglects, of course, the presence of a thin film of the wetting fluid on the pores' surface that are occupied by the second fluid.

Therefore, any of the methods described in Chapter 10 for computing the effective permeability or conductance of pore networks may be utilized for calculating the RPs to the fluid phases. Heiba *et al.* (1982) and Sahimi *et al.* (1986a) developed and implemented the model. Heiba *et al.* (1982) used a Bethe lattice (see Chapters 3 and 10) to take advantage of the analytical formulae for its conductivity, while Sahimi *et al.* (1986a) used a simple-cubic network and computer simulations in order to compute the RPs. Figure 15.1 presents the results obtained with a simple-cubic network. A comparison between Figures 15.1 and 14.8 shows that all the qualitative aspects of the experimental data are reproduced by the model. Note that, as described in Chapter 4, drainage is better described by a bond percolation process, whereas imbibition is more complex and represents a site percolation problem (see below).

Heiba *et al.* (1983) extended their model to the case in which the porous medium is intermediately-wet, or has mixed wettability characteristics, and to the case where there are three fluids in the porous medium, such as, for example, oil, water, and gas (Heiba *et al.*, 1984). Consider the case of an intermediately-wetted porous medium. For such a porous medium, both the primary and secondary displacements are considered as a drainage process. Therefore, the formulae developed by Heiba *et al.* (1982, 1992) for the drainage process can be extended straightforwardly and modified for this case. Heiba *et al.* (1983) showed that their model can predict all the relevant experimental features of the RPs and capillary pressure for intermediately-wetted porous media (see Figure 14.10).

Ramakrishnan and Wasan (1984) used similar ideas and developed expressions for the RPs, and also considered the effect of the capillary number Ca on them. Just

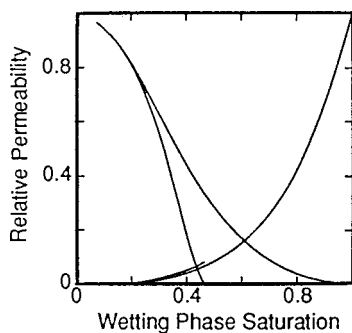


Figure 15.1 Two-phase relative permeabilities, as predicted by the percolation model of Heiba *et al.* (1982, 1992) and Sahimi *et al.* (1986a), using a simple-cubic network. One fluid is strongly wetting while the second fluid is completely non-wetting.

as the residual saturations S_r depend on the Ca (in fact, $S_r \rightarrow 0$ as $Ca \rightarrow \infty$), the RPs also depend on the Ca . Normally, if the capillary number is small, the RPs do not exhibit great sensitivity to the Ca . Evidence for this assertion is provided by the experimental data of Amaefule and Handy (1982). However, as the Ca increases, the RP curves lose their curvature and in the limit $Ca \rightarrow \infty$, they become straight lines. Ramakrishnan and Wasan (1984) developed formulae that took the effect into account. Levine and Cuthiell (1986) used an effective-medium approximation (see Chapter 10) and a percolation model similar to that of Heiba *et al.* to calculate the RPs to two-phase flows in porous media.

15.1.2

Random Site-Correlated Bond Percolation Models

Some have argued that the size distributions of both pore bodies and pore throats must be taken into account. Figure 15.2 presents an example of such distributions. Chatzis and Dullien (1982) used a network model in which the sites represented the pore bodies to which random radii were assigned. The pore throats were represented by the bonds with effective radii that were correlated with those of the sites. Using the model, Chatzis and Dullien (1982, 1985), Diaz *et al.* (1987), and Kantzas and Chatzis (1988) computed the RPs and capillary pressure curves for sandstones. On the other hand, Wardlaw *et al.* (1987) experimentally determined the correlations between the pore bodies and pore throats sizes, and found that there are little, if any, such correlations in Berea sandstones, but that there may be some correlations between the two in the Indiana limestone. Figure 15.2 presents the two size distributions for a Berea network. Note that the throat size distribution appears to be bimodal.

Li *et al.* (1986), Constantinides and Payatakes (1989), and Maier and Laidlaw (1990, 1991b) also proposed network models in which the sizes of the pore bodies and throats were correlated. In spite of the fact that the correlated model is much more detailed than the random bond model, its predictions for the RPs are not fundamentally different from those of the random percolation model.

15.1.3

Invasion Percolation

Invasion percolation (IP) was first proposed by Lenormand and Bories (1980), Chandler *et al.* (1982), and Wilkinson and Willemsen (1983). In the IP model, the network is initially filled with a fluid called the *defender* – the fluid to be displaced. To each site of the network is assigned a random number uniformly distributed in $[0, 1]$. Then, the displacing fluid – the *invader* – is injected into the medium to displace the defender. It does so by choosing at each time step the site next to the interface that has the smallest random number. If the random numbers are interpreted as the resistance that the sites offer to the invading fluid, then choosing the site with the smallest random number is equivalent to selecting a pore with the

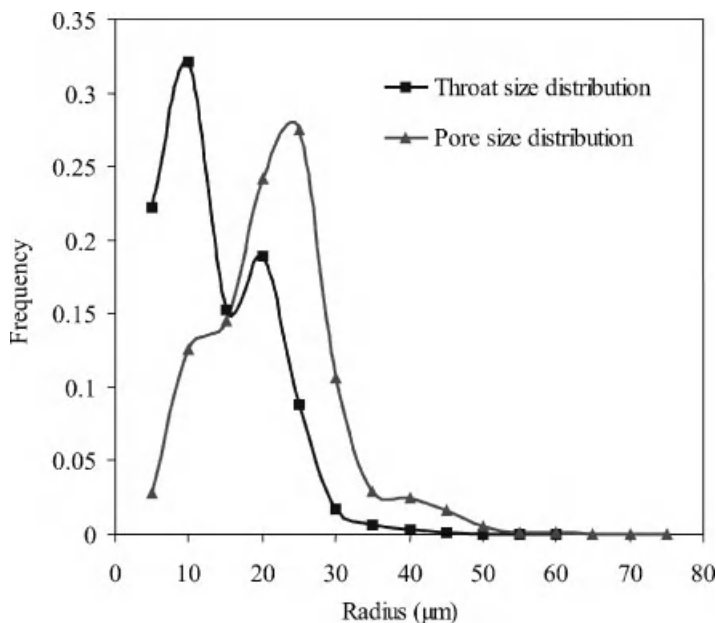


Figure 15.2 Pore and throat size distributions for the pore network equivalent of a Berea sandstone (after Piri, 2003; courtesy of Dr. Mohammad Piri).

largest size and, hence, the IP model simulates the drainage process. A slightly more tedious procedure can be used for working with bonds instead of sites.

A similar IP model can be devised for imbibition, during which a wetting fluid is drawn spontaneously into a porous medium and into the smallest constrictions for which the capillary pressure is large and negative, whereas it enters last into the widest pores. Displacement events are, therefore, ranked in terms of the largest opening that the invading fluid must travel through since it is from the larger capillaries or bonds that it is most difficult to displace the defender. Imbibition is, therefore, a *site* IP, whereas drainage in which the invader has the most difficulty with the smallest constrictions is a *bond* IP.

Two versions of the IP model have been developed. In one model, the defender is incompressible and, therefore, if its blobs (clusters) are surrounded by the invader, they become trapped. This model was studied by Chandler *et al.* and Wilkinson and Willemsen, and is called the trapping IP (TIP). In the second model, trapping is ignored – the displacing fluid displaces an *infinitely-compressible* defender. This version of the IP model – the nontrapping IP (NTIP) – was studied by Wilkinson and Barsony (1984). Note that the IP model represents a dynamical growth process, as opposed to random percolation that is a static model. Figure 15.3 presents the invasion clusters in 2D with and without trapping.

There is a close connection between the IP without trapping and random percolation, first pointed out by Wilkinson and Barsony (1984), who used Monte Carlo simulations to study the models. To see the connection, define an *acceptance profile*

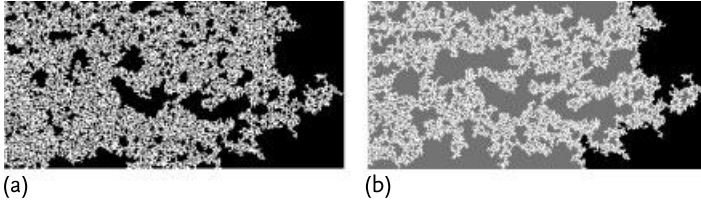


Figure 15.3 Invasion clusters in a two-dimensional system without (a) and with (b) trapping (courtesy of Dr. Fatemeh Ebrahimi).

$a_n(r)$ such that $a_n(r)dr$ is the probability that the random number r selected at the n th step of the invasion is in the interval $[r, r + dr]$. Then, as $n \rightarrow \infty$, one has

$$a_\infty(r) = \frac{1}{p_c}, \quad r < p_c, \quad (15.4)$$

and $a_\infty = 0$ for $r > p_c$, where p_c is the percolation threshold. Monte Carlo simulation of Wilkinson and Barsony (1984) and theoretical analysis of Chayes *et al.* (1985) support Eq. (15.4). Equation (15.4) also provides a precise method for estimating the percolation thresholds in *random percolation* models.

From a conceptual point of view, the IP is perhaps a more appropriate model of capillary-controlled displacements than the random percolation models, with the most obvious reason being the fact that there is a well-defined interface that enters a porous medium from one side and displaces the defender in a systematic and realistic way. Thus, the concepts of history and the sequence of pore invasion according to a physical rule are naturally built into the model, which is also supported by ample experimental evidence.

Lenormand and Zarcone (1985a) displaced oil (the wetting fluid) by air (the non-wetting fluid) in a large and transparent 2D etched network. Their data led to $D_f \simeq 1.82$ for the fractal dimension of the invasion cluster at the breakthrough point (see Eq. (15.5) below), which is consistent with what 2D computer simulations of the IP with trapping yield (see below). Jacquin (1985) and Shaw (1987) also performed experiments that provided strong support to the validity of the IP model. Shaw (1987), for example, showed that if a porous medium, filled with water, is dried by hot air, the dried pores – that is, those filled with air – form a percolation cluster with the same D_f as that of the IP. Stokes *et al.* (1986) used a cell packed with glass beads, an essentially 3D pore space. The wetting fluid was either water or a water-glycerol mixture, while the non-wetting fluid was oil. When the oil displaced the water (drainage), the resulting patterns were consistent with an IP process. Chen and Wada (1986) used a technique in which one uses index matching of the fluids to the porous matrix to “look” inside the porous medium. Their data were consistent with an IP model. Chen and Koplik (1985) used small 2D etched networks with oil and air as the wetting and non-wetting fluids, respectively, and found that their drainage patterns were consistent with the assumptions and results of the IP. Lenormand and Zarcone (1985b) used 2D etched networks and a variety of wetting and non-wetting fluids, and showed that their drainage experiments were completely consistent with an IP description of the phenomenon.

Others have used the IP or a modification of it to explain drying of wet porous media. Shaw (1987) showed that the evaporation of a liquid from a porous medium produces a modified form of the IP. Prat (1995) also studied the similarities and differences between the IP and drying. Tsimpanogiannis *et al.* (1999) carried out experiments with 2D micromodules and pore network simulations to analyze drying fronts in porous media. They concluded that the process is similar to the IP in a gradient (see Section 15.8). For insightful experiments on evaporation from porous media, which may also be modeled as a type of the IP, see Shokri *et al.* (2009, 2010).

15.1.4

Efficient Simulation of Invasion Percolation

Even though the IP model was proposed 30 years ago and was intensively studied in the 1980s, it has received renewed attention over the past 15 years because (1) it is connected with a wide variety of seemingly unrelated problems, and (2) its scaling properties and the structure of the invading clusters have turned out to be far more complex than was previously thought. A good review of such applications is given by Ebrahimi (2010).

Although the IP model is conceptually simple, its simulations, particularly its trapping version – the TIP – is difficult and time consuming. Therefore, the development of an efficient algorithm for the simulation of the IP was of prime importance for a long time. Sheppard *et al.* (1999) and Knackstedt *et al.* (2000) developed a new algorithm for the simulation of the IP that is the most efficient method currently available. Let us describe the algorithm for the TIP.

In the conventional simulation of the TIP, the search for the trapped regions is done after every invasion event using a Hoshen–Kopelman algorithm (see Chapter 3) which traverses the entire network, labels all the connected regions, and then considers only those sites (bonds) that are connected to the outlet face as the potential invasion sites (bonds). A second sweep of the network is then done to determine which of the potential sites (bonds) is to be invaded in the next time step. Thus, each invasion event demands $\mathcal{O}(N^2)$ calculations, where N is the number of sites (bonds) in the pore network. Such an algorithm is highly inefficient for two reasons. (1) Because after each invasion event, only a small local change is made in the interface, implementing the global Hoshen–Kopelman search is unnecessary. (2) It is wasteful to traverse the entire network at each time step to find the most favorable site (bond) on the interface since the interface is largely static.

Sheppard *et al.* (1999) tackled the first problem by searching the neighbors of each newly invaded site (bond) to check for trapping. This is ruled out in almost all instances. If trapping is possible, then several simultaneous breadth-first searches (those that begin at the upstream face) are used to update the cluster labeling as necessary. This restricts the changes to the most local region possible. Since each site (bond) can be invaded or trapped at most once during an invasion, this part of the algorithm scales as $\mathcal{O}(N)$.

The second problem is solved by storing the sites (bonds) on the fluid–fluid interface in a list, sorted according to the capillary pressure threshold (or size) needed to invade them. The list is implemented via a balanced binary search tree so that in-

sertion and deletion operations on the list can be performed in $\log(n)$ time, where n is the list size. The sites (bonds) that are designated as trapped using the procedures are removed from the invasion list. Each site (bond) is added and removed from the interface list at most once, limiting the cost of this part of the algorithm to $\mathcal{O}[N \log(n)]$. Thus, the execution time for N sites (bonds) is dominated (for large N) by list manipulation and scales at most as $\mathcal{O}[N \log(N)]$. In practice, the time and memory requirements depend on the total number of network sites (bonds) and those forming the cluster boundary. For example, it was found empirically that for the 3D TIP, the execution time scales as $L^{1.24}$ (L is the network's linear size), and the memory use is 20 bytes for each network site, plus 64 bytes for each cluster site.

Another problem of interest is identifying the minimum path between two widely-separated sites on the invasion cluster. Sheppard *et al.* (1999) and Knackstedt *et al.* (2000) developed a new optimal algorithm for this purpose. The algorithm consists of three major steps:

1. Using a breadth-first search algorithm, one labels each site in the cluster with its "cluster distance" from the inlet face, and then uses the information to "burn" backwards (see Chapter 3) from the outlet face. At the same time, one constructs the "branch points list" – a list of all the cluster sites that are adjacent to the *elastic* backbone, but are not part of it. The backbone is the multiply-connected part of the invasion cluster, while the elastic backbone is the union of all multiply-connected paths. The branch points list is ordered with the sites closest to the inlet face listed first.
2. The simulation stops if the branch points list is empty. Otherwise, one performs a depth-first search (see Chapter 3) from the last site in the branch points list, flagging all the sites that are visited. During the search, unexplored branch points are added to the branch points list, while another list tracks the sites that have been flagged as visited. One then performs an important optimization during the depth-first search: If there are multiple branches from a single site, the site labeled as being closest to the inlet face is always the first to be explored.
3. The depth-first search terminates when one of two conditions is satisfied: (1) the search contacts the backbone again at a different site from whence it started, in which case the sites in the visited-sites list are flagged as backbone sites, or (2) it retreats back to its starting site, at which point there will be no sites left in the visited-sites list. The algorithm then continues at step two.

15.1.5

The Structure of Invasion Clusters

Similar to the sample-spanning percolation cluster at the percolation threshold, the I_p cluster at the breakthrough point is a fractal and self-similar object. Therefore, the most accurate way of characterizing its structure is through the fractal dimensions of the cluster and its various subsets, for example, the minimum path and the backbone. To estimate the various fractal dimensions, one may employ one of

the two methods. One is based on the scaling of the clusters' or paths' mass with their linear size. For example, for the invasion cluster at the breakthrough point, we must have (see also Chapter 3)

$$M \propto L^{D_f}, \quad (15.5)$$

where M is the mass of the cluster, that is, the number of invaded sites (bonds) in the network, L is the linear size of the sample, and D_f is the fractal dimension.

The second method is based on studying the *local* fractal dimensions and their approach to their asymptotic value as M becomes very large. For example, for the invasion cluster, the local fractal dimension $D_f(M)$ is defined as

$$D_f(M) \equiv \frac{d \ln M}{d \ln R_g}, \quad (15.6)$$

where R_g is the cluster's radius of gyration. According to finite-size scaling theory (FSST) (see Section 3.9), $D_f(M)$ converges to its asymptotic value for large M according to

$$|D_f(M \rightarrow \infty) - D_f(M)| \propto M^{-\omega}, \quad (15.7)$$

where ω is an *a priori* unknown correction-to-scaling exponent (see Chapter 3) and, thus, it must be estimated from the data. Combining Eqs. (15.6) and (15.7), and taking $R_g \propto L$ yields a differential equation with the solution being (Sheppard *et al.*, 1999)

$$c_1 + D_f M^\omega = c_2 L^{\omega D_f}, \quad (15.8)$$

where c_1 and c_2 are constants. Thus, the data are fitted to Eq. (15.8) in order to estimate *both* D_f and ω *simultaneously*. As pointed out in Chapter 3, the choice of ω is crucial to the accurate estimation of the fractal dimensions.

Wilkinson and Barsony (1984) hypothesized that $\Delta = \beta + \gamma = \nu D_f$ is the same for the IP and random percolation models. Here, β , γ , and ν are the standard percolation exponents introduced in Chapter 3. In the literature on percolation theory, Δ is called the *gap exponent* (Stauffer and Aharony, 1994). The hypothesis was consistent with the numerical results of Wilkinson and Barsony (1984). An exact solution of the problem on the Bethe lattice (Nickel and Wilkinson, 1983) also confirmed the hypothesis.

Important differences arise in the structure of the invading fluid paths, depending on whether one considers the NTIP or TIP. While the scaling properties (fractal dimensions and other scaling exponents) of the NTIP are the same as those of random percolation described in Chapter 3, it was believed for a long time that the scaling properties of the TIP in 2D are universal and independent of the network type, and distinct from those of the random percolation. In 3D, the scaling properties of the TIP are the same as those of the random percolation because trapping is too weak to change the scaling exponents. Knackstedt *et al.* (2002) carried out extensive simulation of the TIP in a variety of 2D networks. Their results indicated that, contrary to the common belief, the scaling properties of the 2D TIP model may be network-dependent.

Wilkinson (1986) and Sahimi and Imdakm (1988) derived the power-law laws that the capillary pressure, the RPs, and the dispersion coefficients (see Chapters 11 and 12) follow near the residual saturations (see below). Furuberg *et al.* (1988) studied the probability $P_i(r, t)$ (where $r = |r|$) that a site, a distance r from the injection point, is invaded at time t , and proposed a dynamic scaling for the probability,

$$P_i(r, t) \sim r^{-1} f\left(\frac{r^{D_f}}{t}\right), \quad (15.9)$$

where $f(u)$ is a scaling function with the unusual properties that $f(u) \sim u^{\zeta_1}$ ($u \ll 1$), and $f(u) \sim u^{-\zeta_2}$ ($u \gg 1$), implying that $f(u)$ vanishes at *both* ends because at time t , most of the region within the distance r has already been invaded, and new sites close to the interface that can be invaded are rare. Note that Eq. (15.9) implies that the most probable point at which the advancement of the interface between the two fluids takes place is at $r \sim t^{1/D_f}$. Roux and Guyon (1989) proposed that $\zeta_1 = 1$, and $\zeta_2 = \tau_p + \sigma_p - D_h/D_f - 1$. Here, τ_p , σ_p , and D_f are the standard percolation exponents and fractal dimension (see Chapter 3), while D_h is the fractal dimension of the *hull* – the external surface – of the percolation clusters with $D_h(d = 2) = 1 + 1/\nu = 7/4$, and $D_h(d = 3) = D_f \simeq 2.52$ (ν is the exponent of the percolation correlation length; see Chapter 3).

Laidlaw *et al.* (1988) simulated the IP using two algorithms. One was the usual IP described earlier, while in the second algorithm, the displacing fluid invades *all* the accessible sites of less than a given size. They found that while the fractions of invading fluid in the two algorithms are different (which is expected), their scaling properties are the same. Meakin (1991) studied the IP on substrates with multifractal distribution of bond threshold probabilities (see Chapter 7), and found that the spatial correlations do not change the fractal properties of the IP. However, using extensive simulations, Knackstedt *et al.* (2000, 2001b) showed that long-range correlations of the type that exist in large-scale porous media (see Chapter 5) do change the structure of the invasion clusters. Maier and Laidlaw (1991a) investigated the existence of dimensional invariants (such as B_c defined for random percolation in Chapter 3; see Tables 3.1 and 3.2) in the IP.

15.2

Simulating the Flow of Thin Wetting Films

To make pore network simulation of two- and three-phase flow realistic, simulation of imbibition must take into account the effect of thin wetting fluids on the pores' surface, filled by the non-wetting fluid. The films help the wetting fluid to preserve its continuity in the pore space. Cylindrical pore throat can no longer be used because they cannot support flow of thin wetting fluids on their internal surface. Hashemi *et al.* (1999a,b) described in detail how the flow of thin wetting fluid in pore networks can be simulated, utilizing pore throats with square cross sections. In addition, in the presence of the thin films, it is necessary to carefully define what constitutes a cluster of the wetting fluid. Hashemi *et al.* defined such a cluster as a

set of nearest-neighbor pore bodies – the network’s nodes – that are filled by that fluid and are connected to each other either by thin films that flow in the crevices of the throats, that is, the corner areas on the walls of the throats with square cross sections that connect the nearest-neighbor pores, or by the throats themselves if they are filled by the wetting fluid.

In addition, simulating imbibition entails consideration of several types of displacement mechanisms. One is a pistonlike displacement (flat velocity profile) in the throats. Such a displacement is typically followed by several types of pore filling by the invading fluid, the mechanisms of which depend on the pore’s number of nearest neighbor pores that are already filled with the fluid. For a network of coordination number Z , there are Z pore-filling displacement mechanisms denoted by $\mathcal{D}_0$ to $\mathcal{D}_{Z-1}$ (Blunt and Scher, 1995), which represent filling of a pore with 0 to $Z - 1$ connecting throats that contain the non-wetting fluid (see Figure 15.4). To determine how the displacement proceeds, one needs to calculate the capillary pressure required for each mechanism. In general, $P_c(\mathcal{D}_0) > P_c(\mathcal{D}_1) > \dots > P_c(\mathcal{D}_{Z-1})$. Moreover, the process is limited by the largest radius of curvature necessary to fill the pore, which depends on the number of surrounding throats filled with the invading fluid, and $\mathcal{D}_0$ can occur only if the non-wetting fluid is compressible since in this case, it is this fluid that is trapped in a pore surrounded by throats that are filled by the wetting fluid (see Figure 15.4). Calculating $P_c(\mathcal{D}_i)$ and taking into account the effect of the shapes and sizes of the pores and throats as well as the contact angle is, however, very difficult, and even if it were not, the computations would be prohibitive.

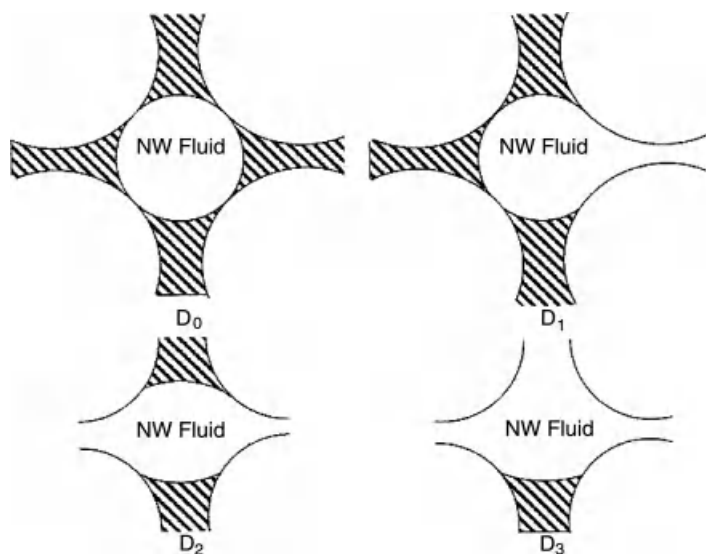


Figure 15.4 Several types of piston-like displacement followed by pore body filling. Shaded areas represent the wetting fluid. Note that in $\mathcal{D}_0$, the non-wetting fluid is trapped inside a pore body (after Hashemi *et al.*, 1999a).

To circumvent the difficulty, a parameterization of $P_c(\mathcal{D}_i)$ is used for describing the advancement of the fluid, which is as follows. The mean radius of curvature R_i for filling by the $\mathcal{D}_i$ mechanism is (Blunt and Scher, 1995)

$$R_i = R_0 + \sum_{j=1}^i A_j x_j, \quad (15.10)$$

where x_j is a random number distributed uniformly in $(0, 1)$, A_j is an input parameter, and R_0 is the maximum size of the adjacent throats. A_j emulates the effect of the pore space variables, and determines the relative magnitude of $P_c(\mathcal{D}_i)$. For example, if for $j \geq 2$, we set $A_j = 0$, then the pore-filling events become independent of the number of the filled throats and, hence, the fluid advance is similar to the IP. If, on the other hand, A_j are relatively large for large values of j , then the model emulates the case of small or medium values of the ratio of pore and throat radii. In the model, R_i , computed by Eq. (15.10), is taken to be the pore radius for the $\mathcal{D}_i$ mechanism of pore filling. Thus, if one fluid is strongly wetting, the critical capillary pressure P_c for pore filling when the pore has i adjacent unfilled throats is taken to be

$$P_c = \frac{2\sigma}{R_i}, \quad (15.11)$$

where σ is the interfacial tension between the two fluids.

During imbibition, one also must consider the snap-off mechanism of throat filling (see Chapter 14): as P_c decreases, the radius of curvature of the interface increases, and the wetting films occupying the crevices of the walls swell. At some point, further filling of the crevices causes the radius of curvature of the interface to decrease, leading to instability and spontaneous filling of the center of the throat with the wetting fluid. The critical capillary pressure for snap-off is given by

$$P_c = \frac{\sigma}{r_t}, \quad (15.12)$$

which is always smaller than $P_c = 2\sigma/r_t$ for pistonlike displacement, implying that snap-off can occur only if pistonlike advance is topologically impossible because there is no neighboring pore filled with the invading fluid.

Therefore, to simulate imbibition and take into account the flow of thin wetting fluid films, consider a pore or a throat filled with the non-wetting fluid, and assume that the wetting fluid is supplied by thin film flow along a crevice of length ℓ . We ignore any pressure drop in the non-wetting fluid and in the portions of the network completely saturated by the wetting fluid (a valid assumption if the capillary number is small), but the pressure drop along the thin wetting layers in the crevices of the network is significant and cannot be neglected. The relation between the wetting fluid flow rate q and the pressure gradient for the thin films in the corners is

$$q = -\frac{r^4}{\beta_s \mu_w} \frac{dP_w}{dx}, \quad (15.13)$$

where μ_w and P_w are the viscosity and local pressure of the wetting fluid, r is the local radius of curvature in the corner, and β_s is a dimensionless conductance factor that depends on the shape of the cross section of the throat and the boundary conditions at the phase boundary. For example (Blunt and Scher, 1995), for square crevices, $\beta_s = 109$ if there is no slip at the interface between the two fluids. More generally, β_s varies anywhere from 15 to 290 (Ransohoff and Radke, 1988). We assume that, locally, the interface is in capillary equilibrium and, hence, $P_w = -P_c = -\sigma/r$, and that flow of the thin films (in the crevices) as well as β_s are independent of the time. Then, for the thin films, $q = \sigma^4/(\mu_w \beta_s P_c^4) dP_c/dx$, that is, flow of the thin wetting films is driven by a capillary pressure gradient in the films. Integrating the equation in $(0, \ell)$, we obtain

$$P_{c0} = P_{cl} \left(1 + 3 \frac{\beta_s P_{cl}^3 q \mu_w \ell}{\sigma^4} \right)^{-\frac{1}{3}}, \quad (15.14)$$

where P_{c0} is the capillary pressure of the element (pore or throat) at the inlet of the crevice, and P_{cl} is the local capillary pressure where the element is being filled – either by pistonlike throat filling or throat filling by snap-off, or by the $\mathcal{D}_i$ mechanism of pore filling.

15.3

Displacements with Two Invaders and Two Defenders

The models of two-phase flow and displacement described so far only involve one displacing fluid – the invader – and one displaced fluid – the defender – whereas in many problems, both at laboratory and field scales, one has a situation in which there are at least *two* invaders and *two* defenders. Recall from Section 14.8, that at the end of the displacement processes described there, the displaced fluid exists only in isolated blobs or clusters of finite sizes that can no longer be displaced by any of the above displacement processes. In order to mobilize and displace such blobs, the Ca must be significantly increased, which would then give rise to three other displacement processes that are quasi-static and dynamic displacement of blobs, both of which are time-dependent phenomena and steady-state dynamic displacement. The last process can be carried out if the displacing and displaced fluids are *simultaneously* injected into the porous medium. Thus, simulating displacements with two invading and two defending fluids is of practical importance. However, such a model is also motivated by other practical considerations.

Recall from Chapter 14, for example, that in a typical experiment for measuring the RPs of two-phase flows, the porous medium is initially saturated with the non-wetting (or the wetting) fluid. Then, a *mixture* (not a solution) of both fluids of a given composition is injected into the sample at a constant flow rate. The two fluids are uniformly distributed at the entrance to the medium, and the flow is maintained until steady state is reached at which point the pressure drop along the sample is recorded that, together with the flow rates of the two fluids and the

Darcy's law, yield estimates of the RPs. Thus, measurement of the RPs involves simultaneous invasion of a porous medium by two immiscible fluids.

As another example, consider two-phase flow in fractures. As described in Chapter 6, a natural fracture usually has a rough self-affine surface. Experimental observations in horizontal fractures indicate (Glass and Norton, 1992; Glass and Nicholl, 1995; Glass *et al.*, 1995) that in two-phase flow through a fracture, when an invasion front encounters a zone characterized on average by much smaller apertures (the zone of the wetting fluid) or much larger ones (the zone of the non-wetting fluid), the invading front advances into that region at the expense of the *already invaded region*. In other words, some apertures in the already invaded region are spontaneously re-invaded by the defending fluid to provide invading fluid for the newly encountered zone. In addition, it has been observed that at low flow rates, gravity-driven fingers drain a distance behind the invading finger tip. Thus, two-phase fluid invasion in horizontal fractures involves *simultaneous* imbibition and drainage of the apertures within the fractures.

Hashemi *et al.* (1998, 1999a,b) developed an IP model with two invaders and two defenders. In simulating such a model, one must recognize that when the fluid clusters are pushed one after another, ℓ in Eq. (15.14) represents the minimum distance between the element to be filled *by the thin wetting films* and the point at the interface between the invading and defending clusters where the path length through the elements completely filled with the wetting fluid is zero. Therefore, when a cluster of the non-wetting fluid pushes a wetting fluid cluster, one has $\ell = 0$, as there is no path of pores and throats that contain thin films of non-wetting fluid in the defender cluster. However, $\ell \neq 0$, if a cluster of the wetting fluid pushes a cluster of the non-wetting fluid since in this case, a thin film of the wetting fluid can participate in the displacement. If each throat is a channel of radius r and characteristic length d (e.g., the distance between two neighboring nodes), we rewrite Eq. (15.14) in dimensionless form using $\ell^* = \ell/d$, $u = q/d^2$, and $P_c^* = P_c r/\sigma$ to obtain

$$P_{c0}^* = P_{c\ell}^* (1 + 3C P_{c\ell}^{*3} \ell^*)^{-\frac{1}{3}}, \quad (15.15)$$

where $C = \beta_s(d/r)^3(u\mu_w/\sigma) = \beta_s(d/r)^3Ca$, with Ca being the capillary number for the flow of thin wetting films. Assuming that the flow rate in each crevice is constant (independent of the time), we consider the filling of an element with the wetting fluid and use Eq. (15.15), replacing ℓ with the minimum distance between the filling element and the inlet of the filled one, where the path length through the throats completely filled with the wetting fluid is zero (i.e., only the paths that consist of the thin wetting films are counted in calculating ℓ).

At time $t = 0$, the network is filled with the non-wetting fluid, and is assumed to be incompressible, so that its trapping by the wetting fluid is possible. A trapped non-wetting fluid cluster can be broken up into pieces only by flow of the wetting films (see below). A site is selected at random on one face of the network for injection, and another site on the opposite face for production. Thus, the boundary condition in the direction of macroscopic displacement is a constant injection

rate. The motion of the injected fluid is represented by a series of discrete jumps in which, at each time step, the invader displaces the defender from the available pores through the least resistance path. In the presence of film flow through the crevices and strongly wetting condition, all the pores are accessible to the wetting fluid, while in the case of no film flow, only the interface pores are accessible. At each stage, the pressure needed for flow of the invader from the injection site to the production site through *all the accessible pores* is calculated by summing up the capillary pressure differences of the pores through *all the possible paths* (see below). The path with the least pressure is then selected. This is completely different from the usual IP models in which only the interface pores are considered. If the wetting fluid is displacing the non-wetting one, then after the element is filled, for every possible new path, the corresponding ℓ is recomputed (in the case of the non-wetting fluid displacing the wetting fluid one has, $\ell = 0$). If the number of filled throats adjacent to any of the pores has increased, the pore filling capillary pressures are updated to represent the proper $\mathcal{D}_i$ mechanism.

Consider, for example, a situation in which the non-wetting fluid is in a cluster that is connected to the inlet of the network, and is in contact with three wetting clusters, which we call WF1, WF2, and WF3. Suppose that the non-wetting fluid tries to displace the wetting fluid. Since there are three wetting fluid clusters that are also in contact with one or several other non-wetting fluid clusters in “front” of them, which in turn touch other non-wetting fluid clusters including one that is connected to the outlet of the network, one must consider all the possible paths of the clusters that are pushing one another, and identify the one that requires the minimum pressure for the displacement from the point of the contact between the inlet non-wetting fluid cluster and the network’s outlet. To do so, one must identify all the non-wetting clusters that are in contact with WF1, WF2, and WF3, which we refer to as the secondary clusters. Then, the tertiary clusters that are in contact with the secondary clusters must also be identified, and so on, until all the possible paths from the inlet non-wetting cluster to the outlet are listed. As such, this is a problem in combinatorial mathematics.

One then calculates the minimum pressure ΔP for displacing a wetting fluid cluster by a non-wetting cluster, and vice versa. Consider, first, the case in which the non-wetting fluid that is connected to the inlet of the network tries to displace a wetting cluster. In this case, only the elements (pores and throats) of the wetting cluster that are at the interface between the two types of clusters are accessible for displacement, as there is no flow of the non-wetting films in any element. Let 1 denote an interfacial element between the invader (the non-wetting fluid) and the defender cluster (the wetting fluid), and 2 denote the outlet point of the wetting cluster to another non-wetting cluster. Recall that the pressure drop between the inlet and outlet elements is $\Delta P_{12} = P_{nw1} - P_{w2}$, and that $P_{nw1} - P_{w1} = P_{c1}$. Therefore, $\Delta P_{12} = P_{w1} - P_{w2} + P_{c1}$. If we assume that there is no pressure drop in the wetting cluster (which is true at low to moderate Ca , then $\Delta P_{12} \simeq P_{c1}$). Thus, we calculate ΔP_{12} for all the interfacial elements between any two non-wetting and wetting clusters that are in contact. Then, for a path that starts from the inlet non-wetting cluster to the outlet of the network, we write the total required pressure

drop $(\Delta P)_{\text{path}}$ as a sum over all such ΔP_{12} s for the non-wetting and wetting clusters that are in contact, being pushed by one cluster on one side (the inlet point 1) and pushing another cluster at another side (the outlet point 2), and belonging to the path:

$$\begin{aligned} (\Delta P)_{\text{path}} &= (\Delta P_{12})_{\text{cluster 1}} + (\Delta P_{12})_{\text{cluster 2}} + (\Delta P_{12})_{\text{cluster 3}} + \cdots \\ &= \sum_i (P_{c1})_{\text{cluster } i} , \end{aligned} \quad (15.16)$$

where cluster 1 is in contact with cluster 2, which is in contact with cluster 3, and so on. In dimensionless form, each term of Eq. (15.16) is calculated using Eq. (15.15). One then selects the path for which $(\Delta P)_{\text{path}}$ is minimum, and keep in mind that when a non-wetting fluid cluster pushes a wetting fluid one, one must calculate the capillary pressure for *all* the interfacial elements (i.e., all the P_{c1}) between the two clusters in order to determine the minimum ΔP_{12} .

Next, consider the case in which the wetting fluid that is connected to the network inlet attempts to push a non-wetting cluster. The general method of selecting the displacement path is the same as before, but two distinct cases must be considered.

1. Suppose that there is no flow of thin wetting films in the crevices. Using the same arguments and notation as above, it is straightforward to show that for any non-wetting cluster that is being pushed by the wetting fluid and is also pushing another wetting cluster, $\Delta P_{12} = -P_{c1}$, assuming again that the pressure in the non-wetting cluster is the same everywhere. The minimum pressure is again determined by considering $(\Delta P)_{\text{path}}$ for all the possible paths.
2. If flow of thin wetting films exists, then the wetting fluid can reach any part of any cluster of the defending non-wetting fluid. One must now calculate the minimum P_c for filling *every* element within the non-wetting fluid cluster, and also those that are at the interface with the wetting fluid. Let one denote an interfacial element between the invading wetting cluster and the defending non-wetting cluster, two be a point inside (a pore of) the non-wetting cluster, and three denote the outlet element of the non-wetting cluster next to another wetting cluster. Since the minimum of such P_c s corresponds to the minimum ℓ (see Eq. (15.15)), then point two is the location of the element within the non-wetting cluster and one is the point at the interface between the wetting and non-wetting fluids that has the smallest distance to that element. Thus, one has $\Delta P_{13} = P_{w1} - P_{nw3}$, and $P_{c1} = P_{nw1} - P_{w1}$ and, therefore, $\Delta P_{13} = P_{nw1} - P_{nw3} - P_{c1}$. Recall that P_{c1} is related to the capillary pressure of the element located at two through Eq. (15.15). Since $P_{nw1} = P_{nw2} = P_{nw3}$, we obtain $\Delta P_{12} = -P_{c1}$. Therefore, in all the cases, the individual ΔP_{12} s only depend on the inlet condition of the clusters, and are independent of the outlet conditions. Note that in both cases, a distinct ℓ is associated with each possible path.

At the end of imbibition, the network is invaded with *both* the wetting and non-wetting fluids, which initiates the fractional flow displacement (FFD). The frac-

tion f_w of the wetting fluid in the injected mixture is fixed. At the early stages of the FFD, there is a continuous path of the wetting fluid through the network, while the non-wetting fluid remains entrapped in its isolated clusters so that the injected wetting fluid can exit from the network's opposite face, whereas the non-wetting fluid accumulates in the network. After successive injections, the injected non-wetting fluid joins its entrapped clusters and, thus, larger clusters of the non-wetting fluid are generated progressively. The invader fluids push many clusters of the non-wetting and wetting fluids, and the wetting fluid clusters that are already in the network.

To determine the path of the mobilized clusters from the inlet to the outlet, we first identify all the clusters and their neighbors. Then, all the possible paths of the clusters from the injection site to the production site are identified and stored in a list. Using Eq. (15.15), the pressure required to mobilize each of the clusters in the list is calculated for each path. The path that requires the least pressure is selected as the flow path between the inlet and the outlet of the network. This represents an important difference between the model for the FFD and the usual IP models in which only the throat with the smallest resistance at the interface is considered. The difference is necessitated by the fact that mobilization of an entrapped fluid cluster is different from simple displacement of one fluid by another in a pore or throat.

Figure 15.5 presents snapshots of the system during three stages of the FFD in a square network for two values of Ca and $f_w = f_{nw} = 0.5$. It is clear that flow of the wetting films plays a major role in the displacement process at low Ca , since the films invade small pores filled with the non-wetting fluid that are far from the interface. Moreover, the effect of the Ca is quite pronounced as the patterns change considerably with increasing the Ca . Interestingly, although due to flow of the thin wetting films during imbibition, the wetting fluid is always sample-spanning and continuous, there is no sample-spanning cluster of the *pores* that are filled with it even at the final stage of the displacement. Since before the FFD begins, the non-wetting fluid was displaced by imbibition, at early stages of the FFD, the clusters of

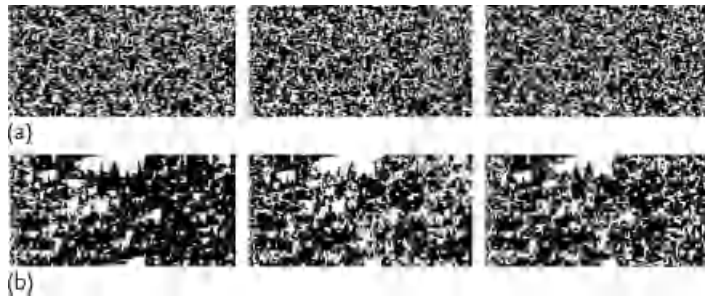


Figure 15.5 Snapshots of the 2D pore space during invasion by two fluids, preceded by imbibition for wetting-phase fractional flow, $f_w = 0.5$, and (a) $Ca = 10^{-5}$ and

(b) $Ca = 10^{-1}$. Black and white areas represent the wetting and non-wetting fluids, respectively. Time increases from left to right (after Hashemi *et al.*, 1999a).

the non-wetting fluid are isolated. However, as the invasion by both fluids proceeds, they become connected progressively and form larger clusters. At some point during the FFD, both fluid phases become continuous – the non-wetting fluid via the sample-spanning cluster of the pores and throats that it occupies, and the wetting fluid through the thin films and the pores that it has invaded. This is a novel feature of invasion by two fluids in 2D with flow of thin films, and is in contrast with the usual IP models in which there is only one continuous fluid phase during the displacement.

15.4

Random Percolation with Trapping

Random percolation with trapping was developed first by Sahimi (1985) and Sahimi and Tsotsis (1985) to model catalytic pore plugging of porous media. In the problem that they studied, the pores of a porous medium plug as the result of a chemical reaction and deposition of the solid products on the surface of the pores. Large pores take a long time to be plugged, and if they are surrounded by small pores that quickly plug, they become trapped and cannot be reached by the reactants penetrating the porous medium from outside.

Accurate computer simulations of Dias and Wilkinson (1986), who proposed the same model for two-phase flow problems in porous media, indicated that most properties of random percolation with trapping in both 2D and 3D are the same as those of random percolation. The pore size distribution (or threshold capillary pressures for pore invasion) that was considered by Dias and Wilkinson was, however, narrow (a uniform distribution in $(0, 1)$). If the pore size distribution is, however, broad (as in the problem studied by Sahimi and Tsotsis), percolation with and without trapping may not necessarily have the same properties.

15.5

Crossover from Fractal to Compact Displacement

As Figure 15.2 indicates, the RP to the non-wetting phase during the primary imbibition by a strongly wetting fluid only vanishes at $S_{\text{rnw}} = 0$, implying that the non-wetting phase is completely expelled from the medium and that the wetting phase *fills up* the pore space. The conclusion is that imbibition is an essentially *compact displacement*. During drainage by a completely non-wetting fluid, however, the RP to the wetting fluid phase vanishes at a *finite* value of S_{rw} , implying that the non-wetting fluid phase does not fill up the porous medium and a fractal cluster emerges. Such differences between imbibition and drainage were already predicted by the random percolation model of Heiba *et al.* (1982, 1992), and was also nicely demonstrated by Lenormand and Zarcone (1984) who used a 2D etched network, injected mercury into the system (drainage), and then withdrew it (imbibition).

The cluster formed during imbibition was totally compact and filled up the etched network.

A definitive study of the problem was made by Cieplak and Robbins (1988, 1990). In their study, the porous medium was represented by a 2D array of disks with random radii where the underlying network was either a triangular or a square network with the disks' centers being at the network's sites. The limit of low capillary number Ca was considered, and the displacement dynamics was modeled as a step-wise process where each unstable section of the interface moved to the next stable or nearly stable configuration. The simulations of Cieplak and Robbins indicated that there are three basic types of instability and the corresponding mechanisms of the advancement of the interface.

1. *Burst*, which happens when at a given capillary pressure P_c , no stable arc connects two disks and, therefore, the interface simply jumps forward to connect to the nearest disk.
2. *Touch*, which happens when an arc that connects two disks, intersects another disk at a wrong contact angle θ , in which case the interface connects to the third disk.
3. *Overlap*, which happens when two nearby arcs overlap. There is no need for the disk to which both arcs are connected and, thus, it may be removed from the interface.

Figure 15.6 illustrates the three mechanisms. To simulate the advancement of the interface, the capillary pressure P_c is fixed and the stable arcs are identified. If instabilities are found, local changes are made to remove them. Then, P_c is increased

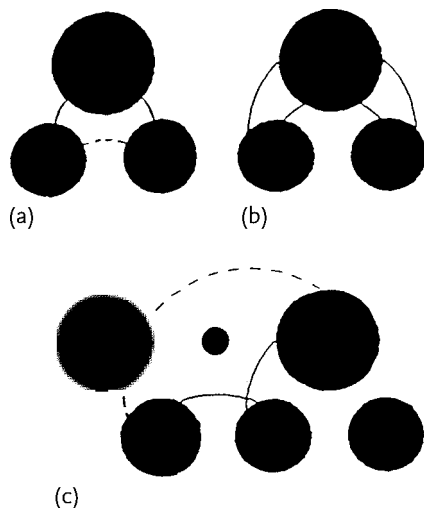


Figure 15.6 Three kinds of growth and instability that occur during an immiscible displacement in a porous medium: Burst (a); touch (b); overlap (c) (after Cieplak and Robbins, 1990).

slightly, the interface is advanced, the possible instabilities are removed again, and so on. As in the TIP, if the invading fluid surrounds a blob (cluster) of the displaced fluid, it is kept intact for the rest of the simulation. If all the disks have the same radius, the resulting patterns are very regular and faceted, and preserve the symmetry of the underlying network, in agreement with the experiments of Ben-Jacob *et al.* (1985).

However, when the radii of the disks are randomly distributed, then the structure of the invasion cluster depends on the contact angle θ . To quantify the effect of θ , we define an interface width w by $w(L) = \langle [h(x) - \langle h \rangle_L]^2 \rangle^{1/2}$, where h is the height of the interface at position x , and $\langle h \rangle_L$ is its average over a horizontal segment of length L . When $\theta \approx 180^\circ$ (i.e., drainage), then the displacement represents an IP and w is of the order of pore size. Cieplak and Robbins (1988, 1990) showed, however, that as θ decreases, the invasion cluster becomes more compact and w increases (see Figure 15.7). At a critical contact angle θ_c , w diverges according to a power law

$$w \sim (\theta - \theta_c)^{-\nu_\theta}, \quad (15.17)$$

where $\nu_\theta \simeq 2.3$. The critical angle θ_c was found to depend on the porosity ϕ of the porous medium; for example, $\theta_c \simeq 29^\circ$ for $\phi = 0.322$, and $\theta_c \simeq 69^\circ$ for $\phi = 0.73$. The exponent ν_θ was found to be universal (independent of the distribution of the disks' radius). The compactness of the cluster for $\theta < \theta_c$ is consistent with the imbibition picture described above.

The divergence of w at θ_c is clearly due to the transition from fractal to compact displacement. For large θ , the interface advances mainly by burst, similar to the IP, and its pattern is independent of θ . However, as $\theta \rightarrow \theta_c$, the overlap and touch incidents become more important, and the interface becomes unstable for almost

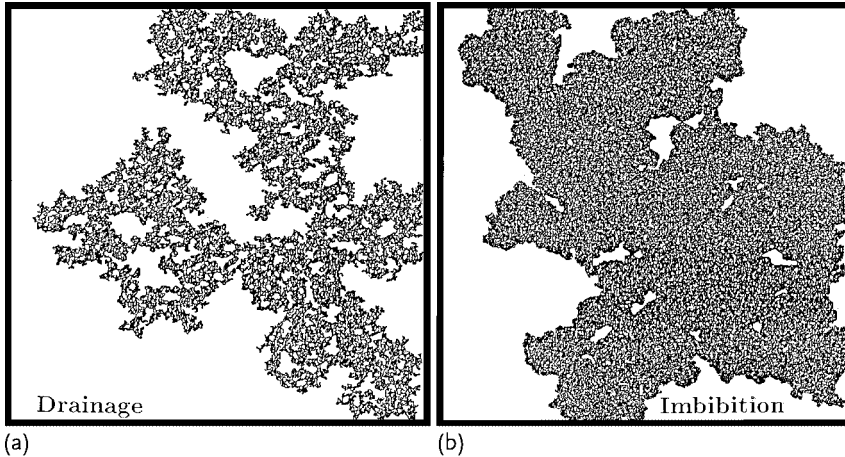


Figure 15.7 Displacement patterns for $\theta = 179^\circ$ (a) and $\theta = 58^\circ$ (b) (after Cieplak and Robbins, 1990).

any configuration of the local geometry. Thus, the growth pattern of the interface changes and, hence, w diverges.

15.6

Pinning of a Fluid Interface

The structure of the fluid interface during imbibition is interesting and different from that during drainage. The invading fluid cluster during imbibition is compact, but capillary forces lead to random local pinning of the interface that results in an interface with a rough self-affine structure demonstrated by the experiments of Rubio *et al.* (1989) and Horváth *et al.* (1991a). The self-affinity of such rough interfaces was first alluded to by Cieplak and Robbins (1988), but was not quantified.

Rubio *et al.* (1989) performed their experiments in a thin (2D) porous medium made of tightly packed clean glass beads of various diameters. Water was injected into the porous medium to displace the air in the system. The motion of the interface was recorded and digitized with high resolution. The experiments of Horváth *et al.* (1991a) were very similar (see below). However, before embarking on an analysis of the results of Rubio *et al.* (1989) and Horváth *et al.* (1991a), let us review briefly the dynamics of rough surfaces and interfaces.

According to the scaling theory of Family and Vicsek (1985) for growing rough surfaces, one has the following scaling form at time t ,

$$h(x) - \langle h \rangle_L \sim t^\beta f\left(\frac{x}{t^{\frac{\beta}{\alpha}}}\right), \quad (15.18)$$

where α and β are two exponents that satisfy

$$\alpha + \frac{\alpha}{\beta} = 2, \quad (15.19)$$

and the scaling function $f(u)$ has the properties that $|f(u)| < c$ for $u \gg 1$, and $f(u) \sim L^\alpha f(Lu)$ for $u \ll 1$, where c is a constant. It is then straightforward to see that

$$w(L, t) \sim t^\beta g\left(\frac{t}{L^{\frac{\alpha}{\beta}}}\right), \quad (15.20)$$

where $g(u)$ is another scaling function and, therefore,

$$w(L, \infty) \sim L^\alpha. \quad (15.21)$$

Note that $w(L, t)$ is a measure of the correlation length along the direction of interface growth. Note also that α is the same as the roughness or Hurst exponent H defined and described in Chapters 5, 6, and 8.

A variety of surface growth models and the resulting dynamical scaling can be described by the stochastic differential equation proposed by Kardar *et al.* (1986)

$$\frac{\partial h}{\partial t} = \sigma \nabla_{\mathbf{r}}^2 h + \frac{1}{2} v |\nabla h|^2 + \mathcal{N}(\mathbf{r}, t), \quad (15.22)$$

where σ is the surface tension, v is the growth velocity perpendicular to the interface, and $\mathcal{N}$ is a random noise. Kardar *et al.* (1986) considered the case in which the noise was assumed to be Gaussian with the correlation

$$\langle \mathcal{N}(\mathbf{r}, t) \mathcal{N}(\mathbf{r}', t') \rangle = 2A\delta(\mathbf{r} - \mathbf{r}', t - t'), \quad (15.23)$$

with A being the amplitude of the noise. For the KPZ model, it has been proposed that (Kim and Kosterlitz, 1989)

$$\alpha = 2(d + 2)^{-1}, \quad \beta = (d + 1)^{-1}, \quad (15.24)$$

for a d -dimensional system. Another stochastic equation was proposed by Koplik and Levine (1985):

$$\frac{\partial h}{\partial t} = \sigma \nabla_{\perp}^2 h + v + A\mathcal{N}(\mathbf{r}, h), \quad (15.25)$$

a linear equation but with a noise that is more complex than that of the KPZ equation. For this model, the numerical work of Kessler *et al.* (1990) indicated that $\alpha(d = 2) \simeq 0.75$.

It is then straightforward to see why pinning of the fluid interface may occur by considering Eq. (15.25) in zero transverse dimension,

$$\frac{\partial h}{\partial t} = v + A\mathcal{N}(h). \quad (15.26)$$

If $v > A\mathcal{N}_{\max}$, where $\mathcal{N}_{\max}$ is the maximum value of $\mathcal{N}$, then $\partial h / \partial t > 0$, and the interface always moves with a velocity fluctuating around v . If, however, $v < A\mathcal{N}_{\max}$, the interface will eventually arrive at a point where $v + A\mathcal{N} = 0$, and is pinned there. Therefore, for a fixed v , there must be a pinning transition at some finite value of A . Indeed, Stokes *et al.* (1988) performed fluid displacement experiments in random packs of monodisperse glass beads in pyrex tubes and measured the capillary pressures at which such a pinning transition takes place.

From their experiments, Rubio *et al.* (1989) found that $\alpha \simeq 0.73$, significantly different from $\alpha = 1/2$, predicted by Eq. (15.24), though consistent with the result of Kessler *et al.* (1990). Horváth *et al.* (1990) reanalyzed Rubio *et al.*'s data and obtained $\alpha \simeq 0.91$, which is larger than all other values. They also carried out their own experiments in a Hele-Shaw cell, packed randomly and homogeneously with glass beads, and displaced the air in the pore space with a glycerol-water mixture, and obtained $\alpha \simeq 0.81$ and $\beta \simeq 0.65$. Although their estimate of α was close to Rubio *et al.*'s as analyzed by Horváth *et al.* (1990), and although the estimates satisfy scaling relation (15.19), they were significantly different from the predictions of Eq. (15.24), but their α is consistent with Kessler *et al.*'s result. Martys *et al.* (1991) employed the model of Cieplak and Robbins (1988) described earlier and showed that below the critical angle θ_c , one has $\alpha \simeq 0.81$, in perfect agreement with Horváth *et al.* (1991a)'s estimate.

How can one explain these results? To our knowledge, no satisfactory explanations have been proposed. Zhang (1990) proposed a modification of the KPZ model

in which the distribution of the noise amplitude is of power-law form

$$\mathcal{P}(A) \sim A^{-(\mu_n+1)}. \quad (15.27)$$

Horváth *et al.* (1991b) then showed that the aforementioned experimental data can be fitted with $\mu_n \simeq 2.7$. A more plausible explanation was proposed by Nolle *et al.* (1993). They calculated the local fluid velocities at the interface and showed that they satisfy a power-law distribution similar to Eq. (15.27) with $\mu_n \simeq 2.7$, in agreement with the estimate of Horváth *et al.* (1991b). Let us also note that Barabási *et al.* (1992) extended the imbibition experiments to three dimensions, and reported that $\alpha \simeq 0.5$.

15.7

Finite-Size Effects and Devil's Staircase

Most of the theoretical discussion so far has been limited to systems that are essentially of infinite extent. If the system is of finite size, the dependence of its macroscopic properties on its linear size L can be investigated using the finite-size scaling described in Chapter 3. So, let us describe the effect of the linear size of a finite porous sample on its capillary pressure and the RPs. Thompson *et al.* (1987b) measured the electrical resistance of a porous medium during mercury injection (drainage) and showed that the resistance decreases (the permeability increases) during the injection process in steps on the so-called Devil's staircase. Their data are shown in Figure 15.8. The steps were irreversible in that small hysteresis loops did not retrace the steps, and were not reproduced on successive injections. When the number $N_{\Delta R}$ of the resistance steps larger than ΔR was plotted versus ΔR , a power-law relation was found,

$$N_{\Delta R} \sim (\Delta R)^{\lambda_R}. \quad (15.28)$$

with $0.57 \leq \lambda_R \leq 0.81$. The magnitude of λ_R presumably depends on the strength of the competition between the capillary and gravitational forces: $\lambda_R \simeq 0.57$ signifies the limit of no gravitational forces, whereas $\lambda_R \simeq 0.81$ represents the limit

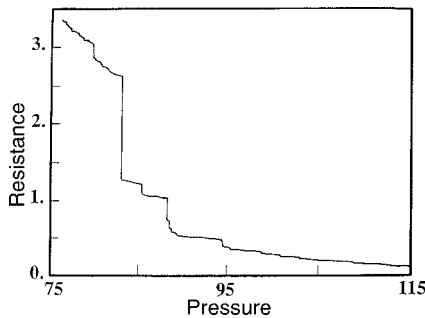


Figure 15.8 Resistance of a sandstone versus the injection pressure during mercury porosimetry (after Thompson *et al.*, 1987b).

in which gravitational forces are prominent. Based on the stepwise decrease of the resistance and the apparent first-order (discontinuous) phase transition (see Figure 15.8), Thompson *et al.* (1987b) concluded that mercury injection should not be modeled by percolation that usually represents a *second-order phase transition*, that is, one that is characterized by a continuous vanishing or divergence of a physical quantity, for example, the permeability or conductivity, as the percolation threshold is approached.

However, simulation of the same process by Katz *et al.* (1988), Roux and Wilkinson (1988), and Sahimi and Imdakm (1988), and related simulation of Batrouni *et al.* (1988) indicated that such a stepwise decrease in the resistance can be predicted by a (random or invasion) percolation. The reason for the stepwise decrease in the sample resistance is that in a *finite sample*, penetration of any pore by mercury causes a *finite* change in the resistance. As the sample size increases, however, the size of the step change decreases, such that for a very large sample, the steps would vanish and the resistance curves become continuous and smooth. In fact, using a percolation model, Roux and Wilkinson (1988) showed that for a 3D porous medium of linear size L ,

$$N_{\Delta R} \sim L^{\frac{3(\mu_p - \nu)}{\mu_p + 3\nu}} (\Delta R)^{-\frac{3\nu}{\mu_p + 3\nu}}, \quad (15.29)$$

where μ_p and ν are, respectively, the critical exponents of the conductivity of the porous medium and the percolation correlation length (see Chapter 3). Thus, $\lambda_R = 3\nu/(\mu_p + 3\nu) \simeq 0.57$, which agrees well with the experimental result in the absence of gravity.

15.8

Displacement under the Influence of Gravity: Gradient Percolation

In all the discussions so far, the effect of gravity on immiscible displacements has been ignored. However, in 3D porous media, the effect of gravity cannot be neglected. The hydrostatic component of the pressure adds to the applied one, which then creates a vertical gradient in the effective injection pressure. Due to the gradient, the fraction of pores that are accessible to the displacing fluid decreases with the height of the system. A modification of the IP model proposed by Wilkinson (1984), and developed further by Sapoval *et al.* (1985) and Gouyet *et al.* (1988), succeeded in taking into account the effect of gravity. However, before describing the models, let us briefly describe a few experimental studies regarding the effect of gravity.

Clément *et al.* (1987) and Hulin *et al.* (1988b) used the following procedure to study gravitational effects. They injected Wood's metal, which is a low-melting point liquid alloy, into the bottom of a vertical and evacuated crushed-glass column. The experiments were carried out at low capillary number Ca by controlling the flow velocity v . After the front reached a given height, the injection was stopped and the liquid was allowed to solidify. The horizontal sections of the front corresponding to various heights were then analyzed, and the correlation function $C(r)$

(see Chapter 4) of the metal distribution in the horizontal planes was determined in order to see whether a fractal structure had been formed. Another series of experiments were carried out by Birovljev *et al.* (1991) in a 2D porous medium. They used transparent models consisting of a monolayer of 1 mm glass beads placed at random and sandwiched between two plates. The system was filled with a glycerol-water mixture, which was displaced by air invading the system at one end.

The competition between gravity and capillary forces is usually quantified by the Bond number Bo, already defined in Chapter 14 and repeated here:

$$\text{Bo} = \frac{\Delta\rho g \ell_g^2}{\sigma}, \quad (15.30)$$

where $\Delta\rho$ is the density difference between the two fluids, g is the gravity, and ℓ_g is the typical size of the grain. Wilkinson (1984) showed that in an immiscible displacement under gravity, the correlation length ξ_g does *not* diverge, unlike in random and invasion percolation that have a diverging correlation length ξ_p (see Chapter 3), but that it reaches a maximum given by

$$\xi_g \sim \text{Bo}^{-\frac{\nu}{1+\nu}}, \quad (15.31)$$

where ν is the exponent that characterizes the divergence of the percolation correlation length ξ_p (see Chapter 3). Thus, $\xi_g \sim \text{Bo}^{-0.47}$, and $\xi_g \sim \text{Bo}^{-4/7}$ in 3D and 2D, respectively. In 3D, there is a transition region where both fluids (displacing and displaced) may percolate in the porous media, the width w of which is given by

$$w \sim \text{Bo}^{-1}. \quad (15.32)$$

Similar results were derived by Sapoval *et al.* (1985) and Gouyet *et al.* (1988) in the context of *gradient percolation*, which is a model in which a gradient G is imposed on the occupation probability p in one direction of the network. The model had, in fact, been considered earlier by Trugman (1983) who called it a *graded percolation*. Sapoval *et al.* and Gouyet *et al.* used scaling arguments similar to Wilkinson's to show that

$$\xi_g \sim G^{-\frac{\nu}{1+\nu}}, \quad (15.33)$$

which is completely similar to Eq. (15.31) in which the Bound number Bo has been replaced with G . The 3D experiments of Hulin *et al.* (1988b) and the 2D experiments of Birovljev *et al.* (1991) were completely consistent with the predictions. For example, Birovljev *et al.* (1991) reported that $\xi_g \sim \text{Bo}^{-0.57}$, where the exponent 0.57 agrees perfectly with the prediction, $\nu/(1 + \nu) = 4/7 \simeq 0.57$.

Wilkinson (1984) also derived an important result regarding the effect of gravity on the residual oil saturation (ROS). He showed by a scaling argument that the difference $S_{ro} - S_{ro}^0$, where S_{ro} is the ROS for $\text{Bo} \neq 0$ and S_{ro}^0 is the corresponding value when $\text{Bo} = 0$ is given by

$$S_{ro} - S_{ro}^0 \sim \text{Bo}^{\lambda_B}, \quad (15.34)$$

where $\lambda_B = (1 + \beta)/(1 + \nu)$, with β being the standard percolation exponent for the fraction of accessible pores near the percolation threshold, or the ROS (see Chapter 3). Thus, Eq. (15.35) predicts that in a 3D porous medium, $S_{ro} - S_{ro}^0 \sim Bo^{0.74}$. In addition, Wilkinson (1984) proposed a simple model for simulating the IP under the influence of gravity.

15.9

Computation of Relative Permeabilities

The two most important properties of two-phase flow in a porous medium are the RPs and capillary pressure. Many empirical correlations have been proposed in the past that relate such properties to a measurable parameter, for example, the fluid saturation. Several equations have also been rigorously developed based on simple models of porous media. However, such equations are as valid as the model used in their development. Two of the models, namely, the sphere pack and bundle of tubes, are too simple – the former is not appropriate for consolidated porous media, while the latter does not take into account the effect of the pores interconnectivity. As a result, the equations derived based on the two models fail to predict the data. Agreement between the theory and data is achieved with such models by inserting adjustable parameters of doubtful physical significance.

Fatt (1956a,b,c) pioneered the pore-network approach by using a regular 2D network to determine capillary pressure and the RPs. However, as described in Chapter 3, it was the work of Larson *et al.* (1977) that ignited the use of pore network models in modeling of two-phase flow in porous media. The paper by Heiba *et al.* (1982) demonstrated how the RPs are accurately computed using pore network models and concepts from percolation theory.

15.9.1

Construction of the Pore Network

It is possible to carry out flow simulations directly on the chaotic pore space by solving the Navier–Stokes equation numerically (Adler *et al.*, 1990, 1992), or by using a lattice-Boltzmann technique (see Chapters 9, 11, and 12). However, such computations can only be done at considerable computational cost. It is, therefore, convenient to construct a pore network that mimics the essential features of the pore space that are relevant to fluid flow. Such network models were already described in Chapters 7, 10, and 11 for single-phase flow and transport. One can reconstruct the network by transforming the reconstructed pore space into a network (Bakke and Øren, 1997).

For predictive modeling, two approaches may be used to characterize the pore network. (1) A simpler approach uses a regular network of capillary elements to represent a system of pores and throats. (2) One tries to model the random topology of the pore space directly by either X-ray computed tomography, a process-based technique, or a statistical technique. The main advantage of regular network mod-

els is their computational simplicity. Not much information is needed to describe the pore system. However, regular networks, although used extensively, have several shortcomings. As already pointed out, cylindrical throats do not support the presence of more than one phase. Perhaps the most fundamental disadvantage of a regular network is that the simulation results may not be directly compared with an analogous physical pore space (Fischer and Celia, 1999; Sok *et al.*, 2002). To make a direct comparison, such parameters as the coordination number and pore size distribution should be tuned to match the “easily” measured data, such as, the capillary pressure, and then the model can be used to predict those properties that are more difficult to measure, for example, the RPs. One may also use irregular networks built based on regular ones (Ghassemzadeh *et al.*, 2001; Ghassemzadeh and Sahimi, 2004a,b), Voronoi networks (Sahimi and Tsotsis, 1997; Fenwick and Blunt, 1998a,b; Blunt and King, 1990, 1991; Dadvar and Sahimi, 2003), Delaunary triangulations (Blunt and King, 1990, 1991) and irregular networks that allow a variable coordination number (Lowry and Miller, 1995). Another approach is to use non-destructive X-ray computed microtomography (Spanne *et al.*, 1994) to image the 3D pore space directly at a resolution of around a micron, which is not, however, sufficient to image the sub-micron size pores that are abundant in carbonates and can only be imaged by such 2D techniques as scanning electron microscopy.

Geometrical properties, for example, the porosity and the two-point correlation function, can be measured from 2D thin sections with high resolution, and used to generate – to reconstruct – a 3D image with the same statistical properties. The advantage of the method is its generality, allowing a wide variety of porous media to be reconstructed. It has been shown, however, that the two-point correlation functions and statistics often fail to reproduce the long-range connectivity of the pore space. One may use multiple-point statistics based on 2D thin-sections as training images in order to generate geologically realistic 3D pore space representations that preserve the long-range connectivity of the pore structure (see also Chapter 7).

15.9.2

Pore Size and Shape

The shapes attributed to pores and throats have a significant effect on the flow and transport properties. In the early pore network models, the pore bodies were either not modeled explicitly at all (they simply connected throats), or assumed to be spherical or cylindrical. However, micromodel experiments (Lenormand *et al.*, 1983) demonstrated that in pores with a rough or angular cross section, the wetting phase may occupy the crevices in layers of order of micron across, and provide extra connectivity (see Section 15.2), while the non-wetting phase occupies the bulk of the pore space. To capture this feature, throats with square or triangle cross sections should be used. One may define a shape factor S_f by

$$S_f = \frac{S}{P^2}, \quad (15.35)$$

where S and P are, respectively, the cross-sectional area and perimeter of the pore.

The irregular triangle is not an exact replica of real pores, though it does have the same range of shape factors as those measured for real porous media. The assumption is that the triangular shape correctly reproduces the balance between the flow in corners (or roughness) and flow in pores' centers. Consider an irregular triangle with corner half angles of α_1 , α_2 , and α_3 , with $0 \leq \alpha_1 \leq \alpha_2 \leq \alpha_3 \leq \pi/2$. α_1 and α_2 are associated with the base of the triangle, and $R = 2S/P = 2PS_f$ is the radius of the inscribed circle. If three rays connect the circle's center to the three vertices, and three lines from the center are drawn perpendicular to the triangle's three bases, then $S = R^2 \sum_{i=1}^3 \cot \alpha_i$. Recalling that, $\alpha_3 = 1/2\pi - (\alpha_1 + \alpha_2)$, one obtains

$$S_f = \frac{1}{4} \left(\sum_{i=1}^3 \cot \alpha_i \right)^{-1} = \frac{1}{4} \tan \alpha_1 \tan \alpha_2 \cot(\alpha_1 + \alpha_2). \quad (15.36)$$

Thus, for such a triangle, S_f varies from zero for slit-like elements to $\sqrt{3}/30 \simeq 0.048113$ for equilateral triangles. A given S_f corresponds to a range of triangles with the limits of the range being $\alpha_{2,\min}$ and $\alpha_{2,\max}$ that, in turn, correspond to the triangles with $\alpha_{2,\min} = \alpha_1 = \alpha$ and $\alpha_{2,\max} = 1/4\pi - 1/2\alpha_1$. Note that the shape factor for rectangular cross sections with all corner half angles being equal to $1/4\pi$ is $1/16$, whereas for circular cross sections, $S_f = 1/4\pi$.

The assumption of an effective circular cross section is reasonable for predicting single-phase properties, and the RP of the non-wetting phase that resides in the centers of the larger pores. For multiphase flow, the capillary entry pressure for non-wetting phase depends on the minimum effective radius along the throat, and for the saturation computation, the pore volumes are essential. Thus, independent expressions for capillary entry pressure, volume and conductance of the network elements must be considered and, consequently, three different radii are important: an inscribed radius to determine capillary pressure at which a non-wetting phase enters the element; a radius that controls the volume, and a hydraulic radius that controls fluid conductance. A variety of pore shapes have been used in the past, including fractal models of roughness (Tsakiroglou and Fleury, 1999a,b; Tsakiroglou and Payatakes, 2000), grain boundary pore shapes (Man and Jing, 2001; Mani and Mohanty, 1998), squares (Blunt, 1998; Fenwick and Blunt, 1998a,b; Hashemi *et al.*, 1998, 1999a,b; Dillard and Blunt, 2000), and triangles (Øren *et al.*, 1998; Hui and Blunt, 2000; Piri, 2003; Piri and Blunt, 2004, 2005a,b).

15.9.3

Quasi-Static and Dynamic Pore Network Models

Two general types of network models may be identified: those for quasi-static displacements that fruitfully use the various percolation models, and the dynamic displacement models. Quasi-static models impose a capillary pressure on the network and calculate the final, static position of all the fluid–fluid interfaces, neglecting dynamic aspects of pressure propagation within the model and the interface dynamics. Dynamic models typically impose a specified inflow rate for one of the

fluids and calculate the subsequent transient pressure response and the associated interface positions. Older pore network simulations for computing the RPs include those of Lin and Slattery (1982), Mohanty and Salter (1982), Sahimi *et al.* (1986a), Blunt and King (1990, 1991), and Jerauld and Salter (1990).

In his model, Wise (1992) removed the throats with the smallest radii from a regular cubic network until the computed capillary pressure (see Chapter 4) matched the data. Rajaram *et al.* (1997) used a network with spatial correlation between the throat radii and optimized both the pore size distribution and the correlation parameters to obtain good agreement with the capillary pressure data. Fischer and Celia (1999) used the same approach, but obtained poor predictions for the RPs. Since the P_c data are not always available, and because the optimal parameters obtained by matching the data and simulation are not necessarily unique (Vogel, 2000), other alternatives have been developed for using the P_c data when statistical properties of a porous medium are mapped onto the network models.

Vogel and Roth (1998, 2001) obtained pore size distribution and pore connectivity from reconstructed samples of unconsolidated soils using multiple 2D thin sections. Hilpert *et al.* (2003) used a similar approach, though by using a cubic network and introducing cross correlation between the throats sizes connected to a single pore. They were able to predict with reasonable accuracy data for both the primary drainage and imbibition capillary pressures for several sphere packs.

Tsakiroglou and Payatakes (2000) combined extracting statistical properties from reconstructed samples with conditioning to capillary pressures from mercury intrusion and retraction. First, the pore size distribution and connectivity information were extracted from thin section analysis with throat size distribution estimated from the experimental capillary pressures. The parameters were then optimized along with the coordination number distribution using a cubic network until a satisfactory match with both intrusion and extraction capillary pressures were obtained. The numerical results for both the capillary pressure and single-phase permeability agreed well with the data.

Sok *et al.* (2002) compared flow and transport properties computed with topologically-equivalent networks to those based on a face-centered cubic. Even when the spatial and cross correlation for the pore and throat sizes as well as the correct distribution of coordination numbers were used, the regular networks yielded poor predictions of both the invasion pattern and the residual saturations. The conclusion was that those parameters that can be obtained from the reconstructed porous medium are not sufficient for reliable predictions of flow and transport properties, and that additional higher-order topological information is needed.

The predicted flow properties not only depend on the network topology and pore size distribution, but also on how each individual pore element is represented and how much pore-scale physics they can accommodate. Most of the network models have used cylindrical tubes to represent the throats. Using a biconical shape rather than a uniform cylinder for throats, Reeves and Celia (1996) argued that more accurate calculations can be carried out. As already pointed out, a major shortcoming

of cylindrical pores is their inability to contain more than one fluid, whereas it is clear that thin films of the wetting layer has a significant effect on flow properties. It was partly for this reason that Hashemi *et al.* (1998, 1999a,b) developed their pore network model that was described in detail in Section 15.3. They used pores and throats with square cross sections.

Kovscek *et al.* (1993) suggested using star-shaped tubes in order to capture the effects of wetting layers, which is also the same cross-sectional shape that was used by, for example, Man and Jing (2001). To incorporate the measured quantities of the highly irregular pore space – cross-sectional area, radius, and perimeter length – into the network model, a dimensionless shape factor G was suggested by Mason and Morrow (1991). A further advantage of such an approach is that not only will it correct for layer volume and conductivity, but that the calculated capillary entry pressure will also be a direct function of the pore shape (Mason and Morrow, 1991; Ma *et al.*, 1996; Øren *et al.*, 1998; Patzek, 2000; Valvatn and Blunt, 2003; Piri and Blunt, 2002, 2004, 2005a,b).

As already pointed out, the wetting physics of a porous medium strongly influences the RPs and capillary pressure and, therefore, including the wetting effect in the pore network model has also been an active research area. The first attempt in this area was undertaken by Heiba *et al.* (1983). More complex is the issue of how mixed wettability (see Chapter 14) is distributed at the pore scale. Kovscek *et al.* (1993) proposed a model whereby the smaller pores become oil-wet while the larger ones remain water-wet. Moreover, visual observation of fluid distribution at the pore scale using cryo- and environmental scanning electron microscopy (Fassi-Fihri *et al.*, 1991; Combes *et al.*, 1998; Durant and Rosenberg, 1998) suggested that the distribution of clay, in particular kaolinite, plays a very important role in determining what parts of the rock are oil-wet.

McDougall and Sorbie (1995, 1997) investigated trends in the RP as well as oil recovery efficiency with a fairly simple network model that did not explicitly include the wetting layers. Dixit *et al.* (1998, 1999, 2000) used the same network models of McDougall and Sorbie (1995) in order to introduce a wettability classification system, relating the recovery from mixed-wet porous media to *aging* and fraction of oil-wet pores. Using the capillary pressures of various flooding cycles, they suggested that it is possible to determine whether oil-wet pores are correlated to the pore size, or are simply randomly distributed.

Blunt (1997, 1998) and Hashemi *et al.* (1998, 1999a,b) used square pores and throats to explicitly model the flow of the thin wetting layers. Using simple geological considerations, expressions for the capillary pressure for various filling events (see Section 15.2) described by Lenormand *et al.* (1983) were developed. In each element (pore or throat), multiple wetting conditions were allowed to exist, with the corners remaining water-wet while the center becoming oil-wet. The effect on the RPs of a wide range of wetting conditions was investigated.

Very limited mixed-wet experimental RP data have been predicted using pore network models (Øren *et al.*, 1998). Jackson *et al.* (2003) used geologically realistic networks of Berea sandstone to obtain the trends in the experimental recoveries.

They assumed that all the invaded pores following the primary drainage became oil-wet. The experimentally measured wettability indices were then matched by varying the distribution of oil-wet contact angles. Øren and Bakke (2003) obtained quantitative agreement with the experimental recoveries, and introduced a method for estimating the oil-wet fraction and the contact-angle distributions using the measured wettability indices. Valvatn and Blunt (2003) used the same approach as that of Jackson *et al.* (2003), including the wetting physics, in their pore network model and obtained very accurate RPs.

The most comprehensive pore network simulator for computing two-phase (as well as three-phase) RPs was developed by Piri (2003), who took into account the effect of contact angle and thin films as well as the connectivity and size distributions of the pores and throats. The results are presented in Piri and Blunt (2004, 2005a,b). Let us then present some of the computed RPs curves and compare them with experimental data.

First, consider the drainage process. The receding contact angle (see Chapter 14) is zero, and the simulations are relatively simple. Figure 15.9 compares the predictions of the pore network model (Piri, 2003) for the RPs of the wetting and non-wetting fluids with the experimental data for Berea sandstone reported by Oak (1990). The data are for oil/water, gas/oil, and gas/water drainage. The agreement between the computed values and the data is excellent.

Next, consider the imbibition process. A uniform distribution of the advancing contact angles (see Chapter 14) is assumed. The initial condition is the irreducible saturation of the wetting fluid, and with all the possible pores and throats occupied by the non-wetting fluid at the end of the primary drainage. As already described in Chapter 14, there is a competition between pore-body filling and the snap-off

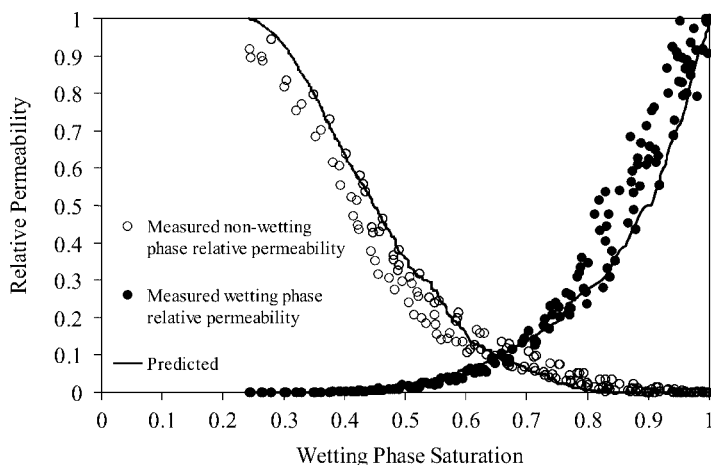


Figure 15.9 Relative permeabilities of oil and water during drainage in Berea sandstone (after Piri and Blunt, 2005b; courtesy of Dr. Mohammad Piri).

events. If the pores are much larger than the throat and the contact angle is low, snap-off is favored, hence leading to a large saturation of the trapped non-wetting fluid. Increasing the contact angle reduces the trapping because the displacement becomes more connected (Jerauld and Salter, 1990; Blunt, 1997). Thus, the range of contact angles was adjusted to be consistent with the trapped non-wetting fluid at the end of imbibition. The ranges for oil/water and gas/oil were found to be $63\text{--}80^\circ$ and $30\text{--}70^\circ$, respectively, consistent with the data of Morrow (1975) described in Chapter 14. Note that all the mechanisms of displacement that were described in Section 15.2 must be implemented during imbibition, including flow of the thin films.

The predicted oil/water RPs during imbibition in Berea sandstone are shown in Figure 15.10 where they are compared with the experimental data Oak (1990). Similarly accurate predictions can be made for the RPs of gas/water and gas/oil systems (Piri, 2003; Piri and Blunt, 2004, 2005a,b). The agreement between the computed and measured RPs is, once again, excellent. Note that if two contact angles are known in a three-phase system, the third contact angle is computed from the equation that relates the three contact angles and the corresponding interfacial tensions:

$$\sigma_{\text{gw}} \cos \theta_{\text{gw}} = \sigma_{\text{go}} \cos \theta_{\text{go}} + \sigma_{\text{ow}} \cos \theta_{\text{ow}} . \quad (15.37)$$

Equation (15.37) follows from the Young–Dupré equation, Eq. (14.1), if we write it once for each of the three fluid pairs and rearrange the three equations. More generally, Blunt (2001) showed that in an n fluid phase system, there are $n(n-1)/2$ contact angles, $(n-1)(n-2)/2$ constraints (such as the Young–Dupré equation), and $(n-1)$ independent contact angles. Finally, non-equilibrium effects in two-phase

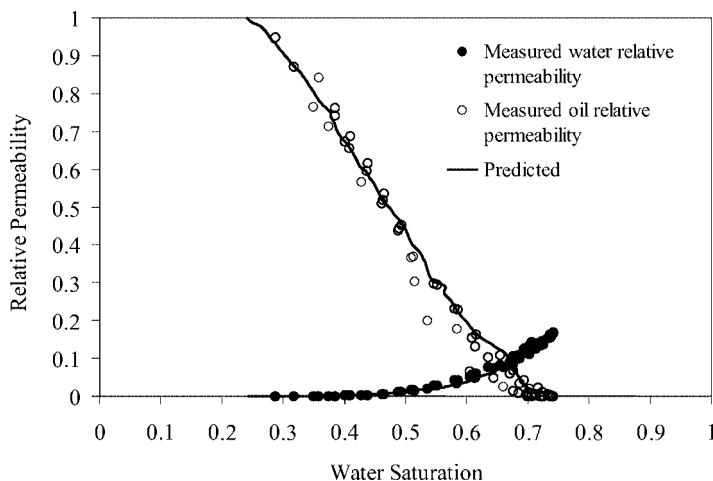


Figure 15.10 Comparison of the measured and computed oil/water relative permeabilities during imbibition in Berea sandstone (after Piri and Blunt, 2005b; courtesy of Dr. Mohammad Piri).

flow in porous media have been studied by a pore network model (Joekar-Niasar *et al.*, 2010).

15.10

Models of Immiscible Displacements with Finite Capillary Numbers

Thus far, we have described capillary-controlled displacements, that is, those in which viscous forces do not play any important role. However, in practice, especially in oil recovery processes, it is often true that for an immiscible displacement process, such as, waterflooding, the capillary number Ca is relatively large so that viscous forces are important. In this section, we describe pore network models of such immiscible displacements with finite Ca numbers (FCN)

The first pore network model of immiscible displacements with FCN was apparently developed by Singhal and Somerton (1977), followed by those of Mohanty *et al.* (1980), and Payatakes *et al.* (1980). In particular, Mohanty *et al.* (1980) used a square network of pore bodies and pore throats with distributed sizes, modeled the displacement of a non-wetting fluid by a wetting one, investigated the effect of pore body and pore throat size distributions, and simulated both low and relatively high Ca regimes.

Detailed, and to some extent quantitative, models of the same phenomena were developed by Koplik and Lasseter (1984, 1985), Dias and Payatakes (1986a,b), Leclerc and Neale (1988), and Lenormand *et al.* (1988). In Koplik and Lasseter's work, the pore space was modeled by a 2D, but non-planar, network of cylindrical pore throats and spherical pore bodies with distributed effective sizes. The local coordination number of the network was randomly distributed. To model the displacement, the equations to solve are those for the pressure field throughout the network, and those for the saturations of the two fluid phases. In a pore throat, the pressure drop ΔP is given by

$$\Delta P = -\frac{1}{g_{p1}} Q + P_c - \frac{1}{g_{p2}} Q, \quad (15.38)$$

where Q is the volume flow rate, and g_{pi} is the single-phase flow conductance of fluid region i . In order to justify use of Eq. (15.38), the basic assumption is that away from the interface between the two immiscible fluids, the flow field in each fluid region is unaffected by the other fluid. Equation (15.38) gives rise to a nonlinear fluid flow problem if it is assumed that the radius of the meniscus between the two fluids and, thus, the capillary pressure P_c , change in some manner as the meniscus passes from a pore body or pore throat into the contiguous pore throat or pore body. The nonlinear problem is converted into a constrained linear one if one assumes that the meniscus stops at the interface during the passing period. The flow in the pore throat is, therefore, zero until the constraints are violated and the meniscus either moves forward into the pore body or back into the pore throat.

If only one fluid is present in a pore body or pore throat, then P_c is, of course, dropped from Eq. (15.38). Hence, using Eq. (15.38) and the fact that for each pore

body the conservation law, $\sum_i Q_i = 0$, must be satisfied, one obtains a set of equations for the pressure at the *center* of each pore body that may be solved by a number of numerical methods. Then, in a time step Δt , a meniscus m with velocity v_m moves a distance $v_m \Delta t$, hence yielding a new fluid distribution, and the procedure is repeated. Koplik and Lasseter assumed that the two fluids have the same viscosity ($M = 1$), and did detailed computations to calculate the rate of change of saturations based on the fluids' fluxes and a knowledge of which fluids are crossing the pore-throat boundaries. At relatively a high capillary number, the viscosity ratio M is expected to have a significant effect, which Koplik and Lasseter's model did not capture. Given the computational limitations of the time, their simulations were restricted to very small pore networks.

The model of Dias and Payatakes (1986a,b) was somewhat more sophisticated than that of Koplik and Lasseter and, at the same time, was simple enough to allow computations with larger networks than used by Koplik and Lasseter. They used a square network of pores having converging–diverging segments with a sinusoidal profile. Such a pore model was first used by Payatakes *et al.* (1980) for simulating blob mobilization and dynamics that were described in Chapter 14. For single-phase flow through a pore, the solution to the flow problem, derived by Tilton and Payatakes (1984), was used according to which

$$Q = \frac{\pi c_0 d_p^3}{4\mu(-\Delta P_1)} \Delta P_{cd}, \quad (15.39)$$

where c_0 is a constant, d_p is the smallest diameter of the pore (at the minimum of the sinusoidal profile), ΔP_1 is a dimensionless pressure drop along the pore (which is a function of d_p) when the flow is creeping and the Reynolds number is unity, and ΔP_{cd} is the pressure drop along the converging–diverging pore. For two-phase flow in the pore, the solution of the problem due to Sheffield and Metzner (1976) was used. For the capillary pressure across the interface, the Washburn approximation (see Chapter 4) was used. Various mechanisms of imbibition similar to those described in Chapter 14 were simulated.

In their second paper, Dias and Payatakes (1986b) simulated mobilization of oil blobs using physical mechanisms that were described in Chapter 14. The calculated quantities included the residual oil saturation (ROS) and the distribution of the blobs. They found that the ROS decreases with decreasing M , even for very small values of the Ca . Moreover, for $M < 1$, the ROS decreased as the Ca did (if $Ca > 10^{-7}$), whereas for $M > 1$, the ROS increased slightly with the Ca in the range $10^{-7} \leq Ca \leq 5 \times 10^{-5}$. However, at even higher values of the Ca , the ROS decreased rapidly as the Ca increased. Their simulations also indicated that a waterflood at finite values of the Ca gives rise to blob populations in which most blobs occupy only one pore body, whereas as $Ca \rightarrow \infty$, large blobs are also formed. Such findings are all in qualitative agreement with the experimental data described in Chapter 14.

The more realistic model is due to Lenormand *et al.* (1988), which is completely similar to that of Leclerc and Neale (1988) that was mentioned in the discussion of miscible displacements in Chapter 13. More details on the work of Leclerc and Neale (1988) are given by Kiriakidis *et al.* (1991). Blunt and King (1990, 1991) also

used a similar model. Whereas both Koplik and Lasseter, and Dias and Payatakes replaced the actual nonlinear flow problem (see above) by a sequence of linear problems, and Lenormand *et al.* (1988) solved the actual nonlinear problem. The porous medium was represented by a network of interconnected pore throats with distributed effective radii. Consider a pore between nodes i and j with radius r_{ij} for which the flow rate Q_{ij} is given by (assuming slow fluid flow)

$$Q_{ij} = \frac{\pi r_{ij}^4}{8\ell\mu_{ij}}(P_i - P_j - P_{cij})^+, \quad (15.40)$$

where ℓ is the pore's length, P_i and P_j are the pressures at i and j , and P_{cij} is the capillary pressure in the pore. The mixture viscosity μ_{ij} was assumed to be given by $\mu_{ij} = 1/2[\mu_2(a_i + a_j) + \mu_1(2 - a_i - a_j)]$, where a_i is the fraction of the pore occupied by fluid i . The plus sign in Eq. (15.40) implies that $Q_{ij} = 0$, so long as $P_i - P_j < P_{cij}$. Due to the constraint, Eq. (15.40) is actually a nonlinear relation between the flow rate and the nodal and capillary pressures. Because the actual nonlinear equations are solved, one can simulate the displacements for any Ca and the mobility ratio M .

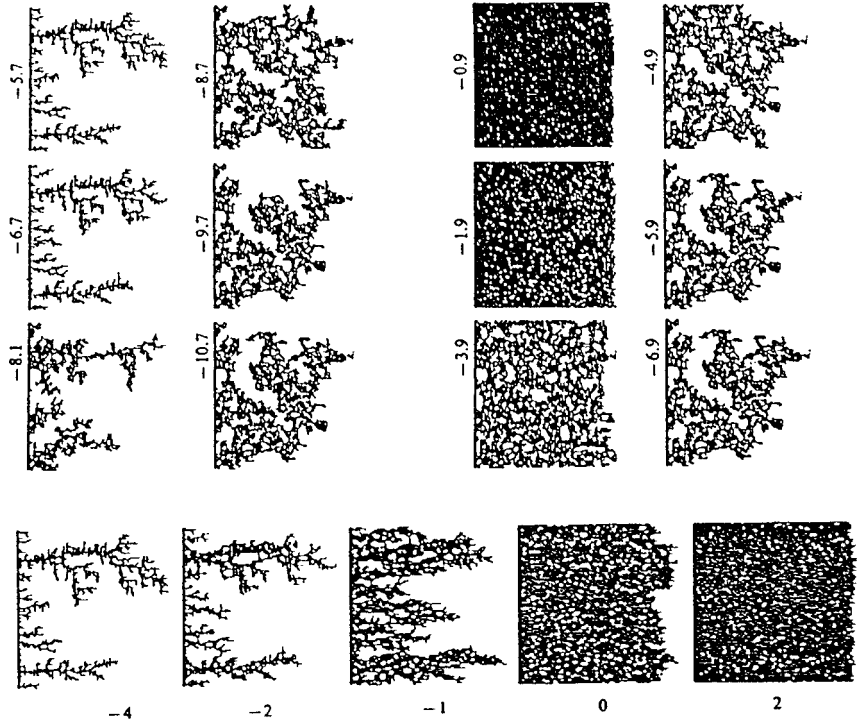


Figure 15.11 Displacement patterns obtained with the model of Lenormand *et al.* Numbers in the horizontal and vertical directions refer to $\log M$ and $\log Ca$, respectively (after Lenormand *et al.*, 1988).

Figure 15.11 presents the displacement patterns computed for several values of the Ca and M . Only the displacement of the wetting fluid by a non-wetting one was studied. Thus, very low values of the Ca correspond to the invasion percolation, whereas very large values of the Ca represent miscible displacements described in Chapter 13. The results are also in excellent agreement with the experiments of Lenormand *et al.* (1983) in 2D etched networks.

The model of Lenormand *et al.* (1988) was significantly generalized by Sahimi *et al.* (1998) in order to take into account the effect of the pore bodies on the displacement pattern. In fact, the pore bodies contain most of the fluid capacity of porous media so that a model that takes into account the effect of the distribution of the effective sizes of both the pore bodies and throats is a more appropriate model. In Sahimi *et al.*'s model, the porous medium was represented by a square network of pores and throats. The size of the throats was distributed uniformly in $(0, 1)$; however, any size distribution could be used. To assign the pore body sizes, consider a pore body i and all the throats that are connected to it. Since its size must be larger than r_{tm} , the size of the largest throat that is connected to it, one takes the size of the pore body i to be $m r_{tm}$, where $m > 1$ is any suitable factor.

Initially, the network is filled with the wetting fluid. The non-wetting fluid invades the network from one face of it by choosing the largest pore throat that is connected to the injection face, which is equivalent to selecting the throat that offers the least resistance to the invading fluid, as the required capillary pressure for the invasion of a throat is the usual, $P_c = 2\sigma/r_t$. Once the throat is filled with the non-wetting fluid, the pore body that is connected to its end is also filled. In practice, the pore body filling is not instantaneous, but such complications have no effect on the results. One then checks all the throats that are available at the new interface between the two fluids, and select again the largest throat for displacement. If a throat, or a cluster of throats, which is filled by the wetting fluid is surrounded by the non-wetting fluid, it remains trapped for the rest of the simulation. The procedure is repeated until the invaded throats form a sample-spanning cluster.

To carry out the flow calculations, one must take into account the following crucial effect. Because the pressure distribution is explicitly calculated, based on which the interface between the two fluids is advanced, and since the pore bodies have finite volumes, one must not only consider the possibility of displacing the wetting fluid from a throat, but also the fact that it may also have to expel the non-wetting fluid from a pore body. To see this, consider a throat which is filled by the wetting fluid, and assume that the two pore bodies that are connected to this throat are both filled by the non-wetting fluid. Sahimi *et al.* (1998) showed that such configurations prevent formation of closed loops. If the non-wetting fluid attempts to enter the throat from one end to expel the wetting fluid, then the wetting fluid will also attempt to expel the non-wetting fluid that is in the pore body at the other end of the throat. Due to this constraint, flow calculation for the model of Sahimi *et al.* (1998) is different from that of Lenormand *et al.* (1988) and, in contrast with theirs, is capable of handling trapping of the clusters of the wetting fluid as well as their mobilization.

Then, consider a throat of radius r_{ij} between pore bodies i and j . If the non-wetting fluid is displacing the wetting fluid, then the volume flow rate in the throat is given by Eq. (15.40) in which P_{cij} is the capillary pressure in the throat between the two fluids, that is, $P_{cij} = P_{nw} - P_w$. If, however, the wetting fluid attempts to displace the non-wetting fluid, then

$$Q_{ij} = \frac{\pi r_{ij}^4}{8\ell\mu} (P_i - P_j + P_{cpj})^+, \quad (15.41)$$

where $P_{cpj} = 2\sigma/r_{pj}$ is the capillary pressure for the pore body j , with r_{pj} being its radius. In Eq. (15.41), $+$ means that

$$Q_{ij} = 0, \quad \text{if } P_{cpj} < P_j - P_i < P_{cij}. \quad (15.42)$$

If constraint Eq. (15.42) is satisfied, then the wetting fluid cannot displace the non-wetting fluid from the pore body. In practice, if the capillary number Ca is small, Eq. (15.42) is always satisfied, implying that the wetting fluid that resides in a throat cannot expel the non-wetting fluid from one of its end pore bodies and, therefore, it is trapped in that throat. The rest of the procedure is similar to that of Lenormand *et al.* (1988).

The most striking result of this model is that at low capillary numbers, the sample-spanning cluster of the non-wetting fluid contains no closed loops. Moreover, there is only one path of the non-wetting fluid from the inlet to the outlet of

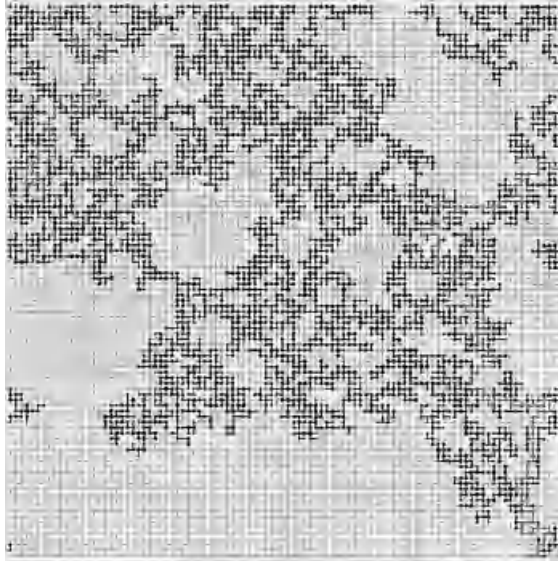


Figure 15.12 The displacement pattern in a 100×100 network. The black area shows the previously invaded area by the non-wetting fluid, dark gray represents the path of the

non-wetting fluid from the inlet (at the top) to the next element to be filled, while light gray shows the wetting fluid. The capillary number is $Ca = 10^{-7}$ (after Sahimi *et al.*, 1998).

the pore space and, therefore, the backbone (the flow-carrying part) of the cluster is simply a long strand. Figure 15.12 shows the displacement pattern in a 100×100 network with $Ca = 10^{-7}$, which contains no closed loops. Moreover, there is a unique path of the non-wetting fluid from the top to the bottom of the pore network. The fractal dimensions D_f and D_{bb} of the sample-spanning cluster and its backbone are $D_f \simeq 1.8$, in agreement with that of the TIP, and $D_{bb} \simeq 1.14$, completely different from $D_{bb} \simeq 1.64$ for 2D percolation (see Chapter 3), and indicative of the strand-like structure of the backbone.

15.11

Phase Diagram for Displacement Processes

Lenormand (1989) studied the crossovers between three regimes of fluid displacements, namely, capillary-controlled displacements – represented by percolation models, and in particular, by the IP – unstable viscous displacements – represented by the diffusion-limited aggregation (DLA) models and their generalizations studied in Chapter 13 – and stable or compact viscous displacements, represented by the anti-DLAs also described in Chapter 13. The result of the study was a phase diagram in the (M, Ca, L) space, where L is the linear size of a porous medium, and M is the mobility or viscous ration. Lenormand (1989) showed that the boundaries of a percolation-type displacement scales as

$$Ca \sim L^{-\frac{\mu_p + \nu + 1}{\nu}} \quad (15.43)$$

towards the stable viscous displacements, and as

$$Ca \sim L^{-\frac{\nu + 1}{\nu}} \quad (15.44)$$

towards the unstable regime. Unstable viscous displacements occur for

$$Ca \sim L^{-1}, \quad (15.45)$$

which extends towards percolation-type displacements as L increases, where μ_p is the critical exponent that characterizes the power-law behavior of the conductivity and permeability of the porous medium near the percolation threshold (see Chapter 3). On the other hand, stable displacements do not depend on the size of the system. Such considerations lead to the phase diagram shown in Figure 15.13.

Fernández *et al.* (1991) also studied the crossover from invasion percolation to a DLA-type displacement. They found that on length scales much smaller (larger) than a crossover length scale L_{co} , invasion percolation (DLA) patterns are obtained. Moreover, according to their scaling analysis,

$$L_{co} \sim \left(\frac{\delta P_c}{Ca} \right)^{\frac{2}{2+D_i}}, \quad (15.46)$$

where δP_c is a measure of the spatial variations of the capillary pressure P_c , and D_i the interface fractal dimension on *small length scales*, which was estimated to

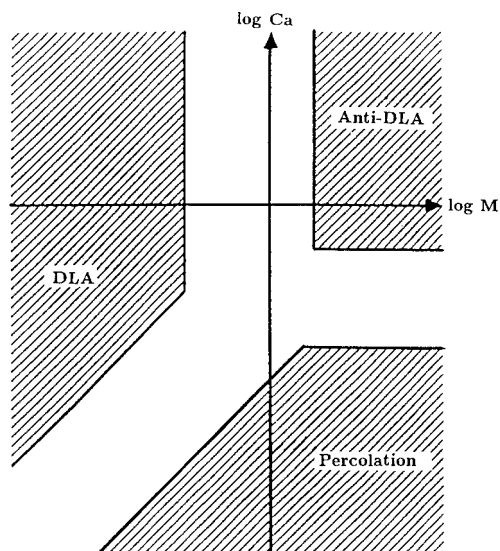


Figure 15.13 Phase diagram for three types of displacements as a function of the capillary number Ca and the viscosity ratio M (after Lenormand, 1989).

be $D_i \simeq 1.3$ in 2D. Xu *et al.* (1998) showed that the phase diagram of Lenormand can be obtained from an invasion percolation model in a gradient studied in Chapter 12; see also Ferer *et al.* (1995, 2003).

15.12

Dispersion in Two-Phase Flow in Porous Media

An important problem in enhanced oil recovery is dispersion in multiphase flows through a porous medium (see, for example, Thomas *et al.*, 1963; Shelton and Schneider, 1975; Salter and Mohanty, 1982; Delshad *et al.*, 1985). The phenomenon is also relevant to groundwater movement in soils that are partially saturated with air (see, for example, Gardner and Brooks, 1957; Biggar and Nielsen, 1960, 1962; Krupp and Elrick, 1968; Gaudet *et al.*, 1977; de Smedt and Wierenga, 1978). In Chapter 11, we described dispersion in single-phase flow through a porous medium. Although there are certain similarities between dispersion in single- and two-phase flows, there are also significant differences between the two. For example, similar to capillary pressure and the relative permeabilities, the longitudinal and transverse dispersion coefficients and the dispersivities in two-phase flow through a porous medium depend on the saturation and the way a given saturation is reached. In other words, the dispersion coefficients during imbibition and drainage are quite different.

If neither fluid phase saturates the porous medium (i.e., the saturation is less than one), then dispersion in a fluid phase in the presence of another immiscible

phase is similar to dispersion in a sample-spanning percolation cluster described in Chapter 11. In this analogy, the fluid phase in which dispersion occurs plays the role of the sample-spanning cluster of the open or occupied pores, and the second phase is similar to the cluster of closed or unoccupied pores. The analogy breaks down if there is significant interphase mass transfer that brings the solute particles from one fluid phase to another. However, we ignore this possibility here since it seems that interphase mass transfer is not very important in most cases.

Thus, reduction of the saturation of a fluid phase (which is similar to reduction of the fraction of occupied bonds in a percolation network) should result in larger dispersivities and dispersion coefficients, and this has indeed been seen in several experiments. Figure 15.14 presents the longitudinal dispersion coefficient D_L renormalized by its value when the wetting phase saturates the system (i.e., at $S_{nw} = 0$) as a function of the non-wetting phase saturation during both drainage and imbibition. Note that during drainage at low values of S_{nw} , D_L is very large and could be divergent at $S_{nw} = 0$. However, in imbibition, D_L first increases as S_w decreases, reaching a maximum, and then decreases as S_w is decreased further. This may be attributed to the existence of thin films of the wetting phase that coat the pore surfaces as a result of which the wetting phase retains its macroscopic connectivity. Thus, although the initial reduction of S_w results in a corresponding reduction in the connectivity of the wetting phase and, thus, an increase in D_L , at low values of S_w , there is a network of thin films of the wetting phase through which the solute particles can be transported in the fluid phase, as a result of which D_L decreases again.

Another important difference between dispersion in one- and two-phase flows is in the velocity-dependence of D_L and D_T during drainage. As long as the saturation of a fluid phase during drainage is not too close to its residual value, the dispersion coefficients pertaining to that phase have a fluid velocity-dependence similar to one-phase flow described in Chapter 11, that is, $D_L/D_m \sim Pe \ln Pe$, where D_m is the molecular diffusivity and Pe the Péclet number. However, as the phase saturation approaches its residual value (the percolation threshold), the volume fraction

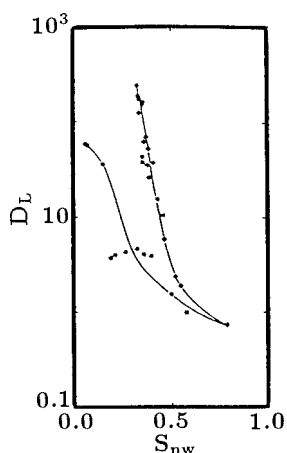


Figure 15.14 The longitudinal dispersion coefficient D_L as a function of the non-wetting phase saturation S_{nw} during drainage (circles), imbibition (squares), and secondary drainage (diamonds). D_L is normalized by its value at $S_{nw} = 0$ (after Salter and Mohanty, 1982).

of the dead-end pores in which the fluid is residing increases. If the time that the solute particles spend in the dead-end pores is significant, there will be a crossover to a new form of the dependence of D_L on the flow velocity given by (see Chapter 11), $D_L/D_m \sim Pe^2$. However, for the new scaling relation to be observed in experiments, the phase saturation must be very close to its residual value so that the volume fraction of the dead-end pores is significant.

Sahimi *et al.* (1982, 1983a, 1986a) extended their pore network model of dispersion in single-phase flow, which was described in Chapter 11, to two-phase flows. The main difference between the simulation of dispersion in one- and two-phase flows is in the distribution of the two immiscible phases. First, one must fix the fluid saturations and distribute the two phases according to one of the models described earlier, for example, the random or invasion percolation model. Next, one determines the flow fields throughout the two fluid phases, which is similar to calculating the flow field in a sample-spanning percolation cluster. Finally, once the flow fields in the two fluid phases are determined, simulation of dispersion in either phase during imbibition or drainage is carried out by exactly the same method described in Chapter 11. Figure 15.15 presents the results of such simulations which have striking similarity with the experimental data shown in Figure 15.14. Since the original simulations of Sahimi *et al.*, other more sophisticated simulation methods have been developed to study the same problem. For example, Bekri and Adler (2002) carried out extensive simulations of dispersion in two-phase flow through porous media by combining three tools that we have described in this book, namely, reconstruction of porous media (to set up the model), the lattice Boltzmann method (for simulating multiphase flow), and random walks (for simulating the dispersion process; see Chapter 11).

The power laws that govern the RPs and dispersion coefficients near the residual saturations or the percolation threshold were derived by Wilkinson (1986) and Sahimi and Imdakm (1988).

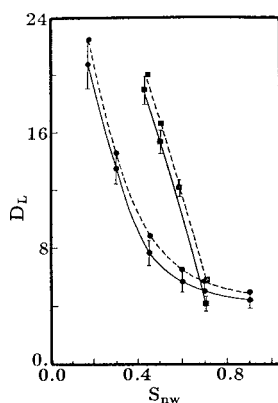


Figure 15.15 Simulation results for the longitudinal dispersion coefficient D_L versus the non-wetting phase saturation S_{nw} during drainage (circles) and imbibition (squares). Dashed curves indicate the simulation results that include diffusion into the dead-end pores (after Sahimi *et al.*, 1986a).

15.13

Models of Two-Phase Flow in Unconsolidated Porous Media

Two-phase flows in packed beds are to some extent similar to flow of two immiscible fluids, that is, oil and water, in consolidated porous media described in Chapter 14 and earlier in this chapter. Therefore, the application of pore network models to modeling such phenomena is natural. The first of such applications was made by Crine *et al.* (1980a,b) who developed a very simple model. One begins with a network in which a randomly-selected fraction p of the bonds are active – they allow fluid flow – and the rest are inactive. It was argued that p corresponds to the irrigation rate or the liquid flow rate. Thus, if p is smaller than the percolation threshold p_c , the pattern of the distribution of the clusters of active bonds corresponds to the rivulet flow (see Chapter 14). However, for $p > p_c$, the pattern is similar to the film flow. As described in Chapter 14, the idea is that in film flow, a sample-spanning cluster of the wetted particles in the packed bed is formed, whereas in rivulet flow, the liquid flow rate is too small to wet all the particles. Figure 14.12 clearly demonstrates the difference between the two flow patterns. To make the patterns look more realistic, Crine *et al.* (1980a,b) converted the random-decorated network of active and inactive bonds to a sort of continuum percolation (see Chapter 3) by drawing circles around the active sites with a radius equal to half of a bond's length. Then, they drew contours of the overlapping circles and “colored” them depending on how many active bonds (per site) were connected to the same site.

Ahtchi-Ali and Pedersen (1986) used a similar method, except that instead of decorating the network randomly, they began at one network face and by moving along a particular direction, selected the active bonds randomly at the interface between the wetted and dry parts of the network. The algorithm is similar to invasion percolation, and is more realistic than that of Crine *et al.* because it simulates, in some sense, the flow of a liquid that enters the packed bed at one face of the system, and leaves it at the opposite face.

Although such percolation algorithms did represent significant advancement over many of the classical models that completely ignored the effect of the morphology of the packed bed, they did not contain enough microscopic physics of the phenomenon to be predictive. More refined and quantitative models were proposed by Zimmermann *et al.* (1987), Chu and Ng (1989), and Melli and Scriven (1991). In the model of Zimmermann *et al.* (1987), a 2D pore network was used to represent a packing of spherical particles. The size of the network was very small and its coordination number was four. Note that, since a bicontinuous structure – a two-phase system in which *both* phases are sample-spanning – cannot exist in 2d (see Chapter 3), one can only simulate the flow of one of the phases (liquid or vapor) if a 2D pore network is used. For this reason, Zimmermann *et al.* (1987) assumed that the gas-phase flow rate was zero, or so small that it did not affect the flow of the liquid. The bonds were assumed to have the same effective radius and, thus, the effect of the pore size distribution of the packing was ignored.

One now needs some rules for how the particles are wetted and how the liquid, once it reaches a node, splits into the outgoing bonds. Zimmermann *et al.* (1987) assumed that when the sum of the flow rates reaching a node (the contact point between two spheres) was less than a critical value, one has partial wetting of the spheres. Otherwise, complete wetting was assumed. At completely wetted spheres, each outflow was assumed to be half the total flow rate arriving there. At partially wetted spheres, the left and right outflows (in the 2D pore network) were assumed to be equal to their inflow counterparts. Zimmermann *et al.* (1987) studied the effect of various factors on the flow of the liquid down the packed bed.

Chu and Ng (1989) used small 3D simple-cubic pore networks in which each bond represented a pore. The packed bed was a regular cubic packing of spheres of equal sizes, but it was tilted in such a way that if viewed from the above, each sphere rests on top of three other spheres spaced evenly apart. Thus, the cubic network was also tilted. Since all the spheres had the same size, the effective radius of all the bonds had to be the same. However, to make the model more realistic, the effective radii of the pores were selected from a distribution function that mimicked the size distribution of a random 3D packing of spherical particles. Due to the 3D character of the network, it was possible to simulate the flow of both phases. Thus, two bicontinuous flow regimes were studied.

Since the upper branch of the hysteresis loop in Figure 14.13 corresponds to liquid film flow over the pore walls and the flow of the gas in the remaining pore space, one may model the phenomenon as an *annular flow* in which a liquid film of a given thickness flows over the walls of the pores of the network, and the gas flows in the bulk in the middle of the pores. Assuming that the inclination angle is χ and that the flow is in the z -direction, and writing down the momentum equation for each phase, we obtain

$$-S_l \frac{dP}{dz} - \tau_{wl} \ell_l + \tau_i \ell_i + \rho_l S_l g \sin \chi = 0, \quad (15.47)$$

$$-S_g \frac{dP}{dz} - \tau_i \ell_i + \rho_g S_g g \sin \chi = 0. \quad (15.48)$$

Here, S_l and S_g are, respectively, the cross-sectional areas for the liquid and the gas, ℓ_l and ℓ_i are the liquid-wall and the interfacial lengths, and τ_{wl} and τ_i are the shear stresses at the wall and at the liquid–gas interface, respectively. One needs correlations that relate τ_{wl} and τ_i to the flow velocities and the fluid properties. As in the case of continuum models described in Chapter 14, such correlations are usually empirical or semi-empirical. However, for laminar flows in tubes, the correlations are derived from the momentum equations. Chu and Ng (1989) used the following well-known equations derived based on the momentum equation (Bird *et al.*, 2007),

$$\tau_{wl} = 2 \frac{\nu_l \ell_l \rho_l}{S_l} v_l, \quad \tau_i = 2 \frac{\nu_g \ell_i \rho_g}{S_i} v_g, \quad (15.49)$$

where ν_l and ν_g are the kinematic viscosities of the liquid and gas phases, respectively.

Thus, the procedure to calculate the upper branch of the hysteresis loop is as follows. For every bond of the network, one uses Eqs. (15.47)–(15.49). At every node of the network, one must have conservation of mass, implying that the algebraic sum of all the flow rates reaching the node must be zero. If one eliminates $\tau_i \ell_i$ between Eqs. (15.47) and (15.48) and uses Eq. (15.49) in the resulting equation, one obtains a single equation for Q_g , the gas flow rate, in terms of dP/dz . Thus, if one writes a mass balance for the gas phase at every node of the network, one obtains a set of linear equations governing the nodal pressures, from the solution of which all the quantities of interest are computed. It is clear that this model is very similar to those described in Chapters 10–12, and the present chapter for flow and dispersion in consolidated porous and fractured media.

The lower branch of the hysteresis loop shown in Figure 14.13 corresponds to *segregated* flow in which a pore is either filled with liquid or gas alone. If both phases co-exist in a pore, they are segregated in the sense that a fraction of the bulk of a pore is filled with the liquid and the rest by the gas. Thus, the same procedure as above with some modest modifications may be used for modeling the lower branch of the hysteresis loop. Figure 14.13 compares the results of such calculations with the experimental data of Christensen *et al.* (1986). The agreement between the computed results and the data is good, demonstrating once again the power of pore network simulation for modeling multiphase flow phenomena in porous media.

Melli and Scriven (1991) used a variation of the model of Chu and Ng (1989) in which most of the volume of the network was assigned to the sites – the pore bodies – rather than the bonds or pore throats. Four flow regimes were also considered: annular (see above), bubbling, flooded (in which the pore throats were filled with only liquid), and *bridged*. The last flow regime occurs in pore throats with constrictions, that is, with slowly varying diverging-converging sections, which exist in packed beds. In such a situation, at high enough liquid flow rates the standing wave below the constriction grows and bridges over, so that only the liquid flows, the gas pressure builds up moderately, and the bridge is pushed down and broken. The process can be repeated again, giving rise to *cycling*.

A more realistic model of fluid flow through packed beds was developed by Bryant *et al.* (1993a,b). They used the Finney packing that Finney (1970) constructed experimentally, and then mapped the pore space of the packing onto an equivalent network. The network is actually one that is constructed on the edges of the Voronoi polyhedra (see Chapter 3). Bryant *et al.* (1993a,b) also corrected for the fact that the pores have diverging-converging segments. Their simulations agreed with the experimental data. A similar procedure was used by Dadvar and Sahimi (2003) who used the same pore network of packed beds to study flow, dispersion and non-linear reaction. Their computed results also agreed with the relevant experimental data. Others, such as Rajaram *et al.* (1997), used pore network models to compute the relative permeabilities for unconsolidated soils.

15.14

Three-Phase Flow

Similar to two-phase flows, extensive experimental investigations, theoretical modeling, and computer simulation studied of three-phase flow in porous media have been undertaken over the past several decades. Let us begin with a brief review of the experimental works.

15.14.1

Measurement of Three-Phase Relative Permeabilities

While, as discussed in Chapter 14, measurement of two-phase relative permeabilities (RPs) is often time consuming, there is just one principle at work: the saturation of one fluid phase increases (decreases) while that of the second fluid decreases (increases). However, measurement of three-phase RPs is particularly challenging because in addition to measuring the saturations, pressure drops and fluxes in the three flowing phases, an *infinite* number of distinct displacement paths also exists since any three-phase displacement involves the variation of *two* independent saturations. Thus, it is impractical to measure the RP for *all* the possible three-phase displacements in a porous medium. However, at the same time, the reality is that the analysis of three-phase displacements is impossible without reliable three-phase RP data.

In the simulation of three-phase flow in large-scale porous media, such as, oil reservoirs, several techniques proposed over the years (Corey *et al.*, 1956; Stone, 1970, 1973; Marek *et al.*, 1991; Robinson and Slattery, 1994; Behzadi and Alvarado, 2010) are used for estimating the three-phase RPs. A good review of the subject was given by Pejic and Maini (2003). In addition, Juanes and Patzek (2004) revisited the foundations of the displacement theory in three-phase flows, and provided general conditions for any RP model to be physically-acceptable anywhere in the saturation triangle of the three phases. However, the reliability of such estimation techniques cannot be tested without extensive experimental data. Relatively speaking though, compared to the two-phase RPs, there is a dearth of experimental studies on three-phase flow in porous media, even though the same factors that affect the two-phase RPs, namely, wettability, viscosity ratio, interfacial tension, injection rate, pore space heterogeneity, and temperature, also affect the three-phase RPs (Akin and Demiral, 1997).

In Chapter 14, we described methods for measuring the RPs. For years, the only widely accepted technique for measurement of the three-phase RPs seemed to be the steady-state method (Maini *et al.*, 1989). The RP data for three-phase flows, measured by this method, have been reported since the 1940s (Leverett and Lewis, 1940) and on through the 1990s (Dria *et al.*, 1993). Typically, 50–100 steady-state measurements are required to construct the RP curve of a given system for one direction of saturation change (Sarma *et al.*, 1994), taking a few days. If hysteresis effects of various directions of saturation changes are also to be evaluated, the task is even more time consuming and difficult.

Such difficulties motivated the development of the faster unsteady-state techniques for measuring three-phase RPs. Beginning perhaps with the work of Sarem (1966) and Donaldson and Dean (1966), three-phase RP data measured by the unsteady-state method have been reported. The works of Nordtvedt *et al.* (1996), Helset *et al.* (1998), and Siddiqui *et al.* (1998) represent some of the more recent measurements. A theoretical analysis of three-phase dynamic displacement experiment was presented by Sahni *et al.* (1996). Piri (2003) provides a comprehensive review. In addition, experiments in micromodels and capillary tubes have been carried out to understand the typical three-phase fluids' configurations at the pore scale, which is crucial to the development of pore network models. However, while the understanding provides a conceptual framework for many three-phase flow phenomena, carefully characterized experiments at the core scale are still necessary for validating the predictions of the pore-scale models, and providing a basis for predicting and interpreting three-phase flow at the field scale.

Three-phase RP data have been reported for water-wet sand packs, water-wet cores, but very limited data for oil-wet cores. Isoperms for gas and water depend mainly on the gas or water saturations, respectively, and are weak functions of other phases present. The permeability to each phase is clearly affected by the saturation history, when there is hysteresis between the imbibition and drainage curves for that phase. The intermediately-wetting phase (oil) appears to be more influenced by the interactions with the other phases. More recent three-phase RP data include the effect of wettability, spreading of the fluids, changes in the hydrocarbons composition, various saturation paths, and the trapping of oil, water and gas in more complex processes, for example, the water-alternating-gas (WAG) processes. Some correlations were proposed by Jerauld (1997), Moulu *et al.* (1999), and Blunt (2000).

15.14.2

Pore-Scale Physics of Three-Phase Flow

To describe and model three-phase flow in porous media, one must pay attention to at least four classes of phenomena and several fundamental quantities which are as follows. Piri (2003) provides a comprehensive discussion.

1. *Spreading coefficients and interfacial tensions*, which were already described in Chapters 4 and 14. The amount of oil that can be recovered following gravity drainage or gas injection has been shown to closely be related to the presence of continuous oil layers, which in turn depends on the capillary pressure and the spreading coefficient (Blunt *et al.*, 1994; Kalaydjian, 1992). The initial spreading coefficient, C_{si} – obtained from interfacial tension of pairs of pure fluids in the absence of the third fluid – and the equilibrium spreading coefficients, C_{se} – obtained from interfacial tension of the three-phase system at thermodynamic equilibrium – can divide a system into three different cases: (1) *non-spreading* with $C_{si} < 0$ and $C_{se} < 0$; (2) *partially spreading* with $C_{si} > 0$ and $C_{se} < 0$, and (3) *spreading* with $C_{si} > 0$ and $C_{se} \approx 0$.

2. *Contact angles and interfacial tensions*, with which we are already familiar. When the oil/water contact angle θ_{ow} of a pore changes with varying oil/water wettability, so also do the remaining contact angles θ_{go} and θ_{gw} . Equation (15.37) imposes a constraint on the three-phase contact angles and interfacial tensions under equilibrium condition. van Dijke *et al.* (2001) presented a linear relationship for determining gas-oil and gas-water contact angles, given the oil/water contact angle and the interfacial tensions,

$$\cos \theta_{go} = \frac{1}{2} \sigma_{go} (C_s \cos \theta_{ow} + C_s + 2\sigma_{go}) , \quad (15.50)$$

$$\cos \theta_{gw} = \frac{1}{2} \sigma_{gw} [(C_s + 2\sigma_{ow}) \cos \theta_{ow} + C_s + 2\sigma_{go}] . \quad (15.51)$$

3. *Wettability alteration and contact-angle hysteresis*: As already described in Chapter 14, based on the oil/water contact angle, porous media are classified into three main groups, namely, water-wet, neutrally-wet, and oil-wet. Direct contact of solid surface with crude oil and subsequent adsorption of the polar components as well as the presence of naturally oil-wet minerals within the pore space may change its original wettability from water-wet to oil wet, which then results in a number of distinct possible fluid configurations during waterflooding. If portions of the pores are oil-wet, water re-enters them as the non-wetting phase and occupies the centers. Oil may also reside as a layer sandwiched by water in the corner and at the center to keep the connectivity of the oil phase and allow for very low residual oil saturations. Kovscek *et al.* (1993) proposed a pore-level model of wettability alteration and fluid configuration. Dixit *et al.* (1998, 1999) introduced a theory that explained hitherto puzzling experimental trends in the recovery in terms of wettability characterized by a contact angle for the oil-wet regions and the fraction of pores that become oil-wet.

As described in Chapter 14, the contact angles also depend on the direction of the displacement. The difference between advancing – wetting fluid displacing the non-wetting one – and receding – the non-wetting fluid displacing the wetting one – contact angles may be as large as 50–90° (Xie *et al.*, 2002), depending on the surface roughness, heterogeneity, swelling, and its rearrangement or alteration by a solvent. Morrow (1975) developed several models for the contact-angle hysteresis by measuring both the advancing and receding contact angles. For each pore and throat, six contact angle can be defined if the wettability is altered: θ_{ow}^A (water displacing oil); θ_{ow}^R (oil displacing water); θ_{gw}^A (water displacing gas); θ_{gw}^R (gas displacing water); θ_{go}^A (oil displacing gas), and θ_{go}^R (gas displacing oil). Other contact angles can be assigned as well for the case for which the pore wettability has not been altered before. For example, for primary drainage, one may define θ_{ow}^{PD} (oil displacing water) and θ_{gw}^{PD} (gas displacing water). In cases that the contact angles need to be calculated from some correlations and it is not clear as to which advancing or receding contact angle is to be used, one should note that the receding contact angle for each fluid phase is less than or equal to the advancing value.

4. *Spreading and wetting layers*: In a typical arrangement of three fluid phases in a water-wet non-circular element, water fills the corners and preserves the continuity of the water phase, the gas occupies the center of the element, and then there is the possibility of oil remaining in a layer sandwiched between the gas and water. These are called the *spreading layers*. The stability of the layers is linked to the spreading coefficient, contact angles, corner angles (configuration of the pore cross section), and the capillary pressures.

In a spreading system ($C_s = 0$), oil spontaneously forms layers between water and gas in the pore space. For non-spreading systems ($C_s < 0$) the oil layers can also be present, but they tend to be stable for a more restricted range of the capillary pressures. The spreading layers are typically a few microns in thickness and have a non-negligible hydraulic conductance, and maintain phase continuity. In contrast, films that were reported by Dullien *et al.* (1989) in smooth bead packs are only a nanometer in thickness, stabilized by molecular forces, and have negligible conductivity.

15.14.3

Pore Network Models

The same steps that were described earlier for modeling of two-phase flow in porous media by pore network models must also be taken for three-phase flows. In addition, one needs to describe each displacement mode separately. We know that if the displacement is a quasi-static process, it is controlled entirely by the capillary pressure. This can then be represented by the replacement of one phase by another at the center of the pore, or the collapse or formation of a layer in a single corner. As described earlier, a capillary pressure is also associated with the transition from one configuration to another.

1. *Primary drainage*: Initially, the network is saturated with water and is strongly water wet, $\theta_{ow}^{PD} = 0$, while all the capillary pressures are zero. Then, oil enters the network, representing migration into an oil reservoir. Then, the invasion percolation algorithm is applied for simulating the process. Mason and Morrow (1991) derived a general expression in terms of the shape factor S_f for the drainage threshold capillary pressure in strongly water-wet triangular pores. Their analysis was extended to include the effects of contact angle and contact-angle hysteresis in equilateral triangular pores (Morrow, 1990), and then for a general triangular pore (Øren *et al.*, 1998). As already pointed out in Chapter 14, the threshold capillary pressure P_c is governed by both the pore shape and the receding contact angle θ_R , and is written in the following general form, already mentioned in Chapter 4,

$$P_c = \left[\frac{\sigma_{ow}}{r} (1 + 2\sqrt{\pi} S_f) \cos \theta_R \right] G_d(\theta_R, S_f, \alpha), \quad (15.52)$$

where r is the inscribed radius of pore or throat, G_d is a dimensionless correction factor for the wetting fluid that might be retained in the corners, and the rest of the notation is as before. In general, G_d depends on the particular corner angles and is not universal for a specific S_f . It has been determined for a variety of pore shapes; see Piri (2003) and Piri and Blunt (2004, 2005a,b). Note that for strongly water-wet porous media ($\theta_R = 0$), $G_d = 1$. Drainage is completed when a target capillary pressure or saturation is reached, or after all the pores and throats have been invaded by oil.

One must then take into account the wettability alteration. When oil initially invades a water-filled pore body or throat, a stable water film protects the pore surface from wettability change by adsorption. At a critical capillary pressure, the film collapses to form a molecularly-thin film, allowing the surface active components in the oil to adsorb on the pore surface. The capillary pressure at which the water film ruptures depends on the curvature of the pore wall and on the shape of the disjoining pressure isotherm. Kovscek *et al.* (1993) presented a detailed analysis of the phenomenon for star-shaped pores, and Blunt (1997) extended the analysis to square pores and presented a parametric model for the critical capillary pressure at which the water film collapses. Distinct advancing oil/water contact angles θ_{ow}^A may be assigned to each oil-filled pore and throat after the primary drainage selected from a specified wettability distribution.

After the primary drainage ends, the model can simulate any sequence of water, gas, and oil invasion. Then, water re-enters the network in a waterflooding process by decreasing the capillary pressure. For the filling process, assignment of the local threshold pressures is complex, as they depend not only on the contact angles and pore space morphology – corner angles and radius of the element of interest – but also on the fluid configuration since penetration of a meniscus of a wetting fluid into a pore from a throat can change the effective radius of the pore. Thus, five additional mechanisms must be considered.

2. *Piston-like filling in throats* refers to the displacement of one phase by another in the center of a throat by a fluid residing at the center of a neighboring pore. The computation of relevant filling capillary pressure is more involved than in drainage because it involves invading a throat that also contains water in the corners that swell as the water pressure increases. If there is no contact-angle hysteresis, then the threshold capillary pressure of imbibition is the same as that of drainage. In a more realistic scenario, the advancing contact angle θ_A is different from the receding contact angle θ_R , which is always true for mixed-wet systems for which $\theta_A \gg \theta_R$. Thus, there is a range of capillary pressures in which the invading interface remains pinned (see the earlier discussion of interface pinning). As the relevant capillary pressure is reduced, each interface remains fixed in the last position obtained during primary drainage, and the contact angle adjusts to a new value called the *hinging contact angle* θ_h with $\theta_R < \theta_h < \theta_A$ in order to maintain capillary equilibrium. θ_h is a function of

the capillary pressure and is given by

$$\theta_{ow}^h = \cos^{-1} \left[\frac{r_{ow}^d}{r_{ow}^{im}} \cos(\theta_{ow}^R + \alpha) \right] - \alpha, \quad (15.53)$$

where α is the corner half angle (assuming that all the corner angles are equal), and the superscripts denote imbibition and drainage.

The wetting phase enters the element after the capillary pressure is lowered sufficiently such that θ_A is attained. During spontaneous (positive capillary pressure) piston-like displacement, the capillary entry pressure is again determined by calculating the force acting on the interface. If all the θ_h for the pores have attained θ_A , the expression for the capillary entry pressure will be the same as that of primary oil flooding with the θ_R replaced by θ_A . If one or more of the pore-scale interfaces have attained θ_A (but not the rest), their corresponding positions are adjusted. Spontaneous displacement might occur for $\theta_A > 90^\circ$. During forced water invasion (negative capillary pressure), the absolute entry pressure is given by Eq. (15.52) with θ_R replaced by $\pi - \theta_A$; see also van Dijke *et al.* (2007).

3. *Pore-body filling* refers to the displacement of one phase at the center of a pore body by movement from the center of adjoining throat(s). The threshold capillary pressure in drainage is similar to the piston-like advancing, but the complexity here for imbibition is that the critical radii of curvature for the pore body filling depend on the number of adjacent throats that are already filled with the invading fluid. For a pore body of coordination number Z , there are Z possible events, $\mathcal{D}_0$ to $\mathcal{D}_{Z-1}$, which represent filling of the pore body with 0 to $Z - 1$ connecting throats that contain the invading fluid. How the displacement proceeds was already described in Section 15.2.
4. *Spontaneous snap-off* corresponds to an event in which the fluid at the center of a pore body or throat is displaced by one residing in the corners or layers. As described in Chapter 14, snap-off occurs when the water layers in the corners swell until the layers in two corners meet, and there is no longer an oil/water-solid contact. At this point, oil/water interface is unstable and the pore body or throat spontaneously is filled with water. Snap-off occurs at a positive capillary pressure (a water pressure lower than the oil pressure) and is possible only if $\theta_{ow}^A < 1/2\pi - \alpha_{\min}$, where $\alpha_{\min}$ is the smallest half angle of the corners in the element. The capillary pressure at which this occurs depends on whether one or several menisci have begun to advance along the pore body wall. It could be that two or more menisci have begun to move, or that only the meniscus in the sharpest corner has begun to advance. In any event, the event with the highest capillary pressure will be the one that occurs because the hinging angles depend on the prevailing capillary pressure.
5. *Forced snap-off* is analogous to spontaneous snap-off, but it is a forced process that occurs at a negative capillary pressure. As the water pressure increases, the curvature of the oil/water interfaces in the corners change according to, $P_i - P_j = 2\sigma_{ij} \cos \theta / r$. However, the oil/water-solid contact cannot move until

$\theta = \theta_{ow}^A$. This means that the curvature varies with the capillary pressure, but the oil/water-solid contact remains in the same place – the interface is pinned – with the contact angle varying as a function of capillary pressure. If the condition $\theta_{ow}^A < 1/2\pi - \alpha_{min}$ is not satisfied, the oil/water interface will bulge out into the oil (the capillary pressure will be negative) until $\theta = \theta_{ow}^A$, and the contact begins to move. Any movement of the contact angle tends to make the local capillary pressure less negative, hence drawing in water from nearby and resulting in the spontaneous filling of the element with water. Snap-off is not favored over a piston-like or pore-body filling event, if there is a neighboring element with the invading phase at the center that is able to carry out the displacement. As already described in Chapter 14 and in Section 15.2, there is a competition between pore-body filling and snap-off during imbibition. In fact, the ratio of pore body and throat radii controls the snap-off process.

6. *Layer formation and collapse:* layer formation is possible by (1) allowing the displaced phase to remain as layer(s) sandwiched between the fluids in the corner(s) and the element's center if the pertinent contact angles, capillary pressures, and corner half angles permit it in piston-like and snap-off events, and (2) by displacement from fluids residing in the layers or center of neighboring elements. The layers may collapse by an increase in the pressure of the fluids on their either side. One should note that some displacements, such as piston-like, pore-body filling and snap-off, change the configurations in *all* the corners of the element, whereas others, for example, layer collapse and formation, change it only in one corner.

15.14.4

Simulation of Three-Phase Flow

Similar to two-phase flow in porous media, since the early 1990s, considerable efforts have been devoted to developing realistic pore network models for three-phase flow, for example, the flow of oil, water, and gas, in porous media. The first of such models was, however, developed by Heiba *et al.* (1984) who extended their percolation model for two-phase flow, described earlier, to three-phase flows. Soll and Celia (1993) developed a capillary dominated two- and three-phase model to simulate capillary pressure-saturation relationship in a water-wet regular porous medium using 2D and 3D pore networks. Hysteresis was modeled by using advancing and receding values for contact angles for each pair of fluids (see Chapter 14). Each pore was able to accommodate one fluid at a time as well as the wetting thin film. Viscous forces were neglected, but the gravity was included.

Øren *et al.* (1994) studied displacement mechanisms during immiscible gas injection into the waterflood residual oil in a strongly water-wet porous medium using a square pore network with rectangular throats and spherical pore bodies in order to compute oil recovery in spreading and non-spreading systems (see below). They did not calculate the RPs, but described a double drainage mechanism where-

by gas displaces the trapped oil that displaces water, allowing the immobile oil to become connected that boosts oil recovery.

Pereira *et al.* (1996) and Pereira (1999) developed a dynamic 2D network model for drainage-dominated three-phase flow in strongly water- and oil-wet porous media when both the capillary and viscous forces are important. They did not calculate the RPs, but generalized the two-phase displacement mechanisms and used throats with lenticular cross sections that allow the wetting and spreading layers to be present. As in Øren *et al.* (1994), a large difference between recoveries in the spreading and non-spreading systems was reported for the water-wet cases due to the existence of oil layers in the spreading systems. Moreover, a reduction in the initial oil saturation for the tertiary gas injection decreased the oil recovery in both spreading and non-spreading systems, indicating that the recovery of intermediate wetting phase is a strong function of the saturation history, whereas the same is not the case for wetting fluid.

Paterson *et al.* (1997) developed a percolation model for a water-wet porous medium to study the effect of the spatial correlations in the pore size distributions on three-phase RPs and the residual saturations. The model assigned the same volume and conductivity to all the pore throats so the fraction of the sites occupied by a fluid phase was the same as its saturation. The simulation indicated lower residual saturations when compared with the uncorrelated ones, while the RPs, when the flow was parallel to the bedding layers, were higher than perpendicular case. Fenwick and Blunt (1998a,b) used a simple-cubic network to model three-phase flow in a porous medium. The cross sections of the throats were equilateral triangles or square in order to simulate strongly water-wet systems. Double drainage was generalized to allow any of six types of double displacement where one phase displaces a second fluid that displaces a third, as observed by Keller *et al.* (1997). The model was able to simulate any sequence of oil, water and gas injection. Using a geometrical analysis, a criterion for the stability of the oil layers was derived that was dependent upon oil/water and gas/oil capillary pressures, contact angles, equilibrium interfacial tensions, and the corner half angle. They were the first to estimate the conductance of an oil layer, which then was used to compute the oil RP. They also proposed an iterative methodology that coupled a physically-based pore network model with a 1D three-phase Buckley–Leverett simulator (see Chapter 14) in order to determine the correct saturation path for a given process with known initial condition and injection fluid.

Mani and Mohanty (1997, 1998) used a cubic network to simulate both dynamic and quasi-static three-phase flow in water-wet systems. Pore bodies and throats were assumed to be spherical and cylindrical, respectively. The model contained two important features: (1) dynamic simulation of capillary-controlled gas invasion for which it was assumed that each fluid phase pressure was not constant across the network, and (2) re-injection of the produced fluids at the outlet of the medium to the inlet in order to simulate larger systems, which was used to study whether the trapped oil ganglia are reconnected by double drainage to form spanning clusters. The model included flow through the wetting and spreading layers with affixed conductance assigned to oil layers. Gas invasion was modeled by three displace-

ment mechanisms, namely, direct gas/water, direct gas/oil, and double drainage. For each displacement, a potential was considered that was the difference between the pressures of the two fluids involved, minus the threshold capillary pressure of the displacement. The displacement with the largest potential was carried out first. Re-injection of the fluids was simulated by replacing the fluid distribution in the inlet zone by the fluids in the outlet region. The simulations indicated that the final oil saturation in spreading system was zero, and the capillary pressure curves for the dynamic and quasi-static simulations were virtually identical.

Laroche *et al.* (1999) developed a pore network model to predict the effect of the wettability heterogeneities with various patterns and spatial distributions on the displacement mechanisms, sweep efficiency, and the fluid distribution in gas injection into oil and water. A dalmation type of wettability heterogeneity was used with continuous water-wet surfaces enclosing discontinuous regions of oil-wet surfaces or vice versa. Saturations, conductances, and the RPs were calculated using techniques similar to those of Fenwick and Blunt (1998a,b).

Larsen *et al.* (2000) used a cubic pore network of pores with square cross section to model three-phase flow in a water-wet porous medium, and simulate a series of micromodel experiments of the WAG injection. Three WAG injections with several gas/water injection ratios were carried out. Van Dijke *et al.* (2002) and van Dijke and Sorbie (2002) developed a regular 3D pore network model of porous media in which each element was allowed to have a different oil/water contact angle in order to simulate wettability heterogeneity. While thin layers of fluids were not explicitly incorporated in saturation or conductance computations, they were allowed to establish the continuity of the fluid phases.

In an attempt to address the full range of possible configurations in mixed wet systems, Piri and Blunt (2004, 2005a,b) presented a 3D pore network model to simulate two- and three-phase capillary dominated processes. Their network model has three essential components: (1) a description of the pore space and its connectivity that mimics real systems; (2) a physically-based model of wettability alteration described earlier, and (3) a full description of fluid configurations for two- and three-phase flow. They used the technique developed by Bakke and Øren (1997) to reconstruct a 3D void space and then converted it to a pore and throat network model. The important mechanisms of immiscible flow at the pore scale that were described in the last section were all included in the model. In all, they analyzed *thirty* generic fluid configurations. Double displacement and layer reformation were implemented, as were direct two-phase displacements and layer collapse events. Thus, the model developed by Piri and Blunt represents the most complete pore network model of three-phase flow that we are aware of. An important aspect of their work was their saturation-tracking path algorithm, the predictions of which reproduced the same displacement path observed in experiments. An example is shown in Figure 15.16, which is compared with the data of Oak (1990).

In general, the predictions of Piri and Blunt for the RPs of gas and water phases were accurate. Figure 15.17 compares the computed three-phase water RP during

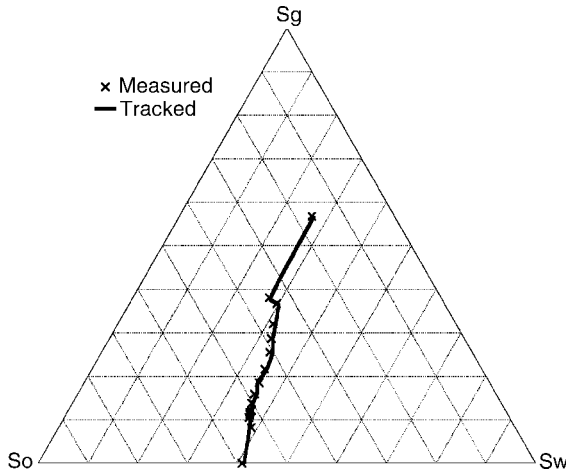


Figure 15.16 Comparison of measured and computed saturation paths (after Piri and Blunt, 2005b; courtesy of Dr. Mohammad Piri).

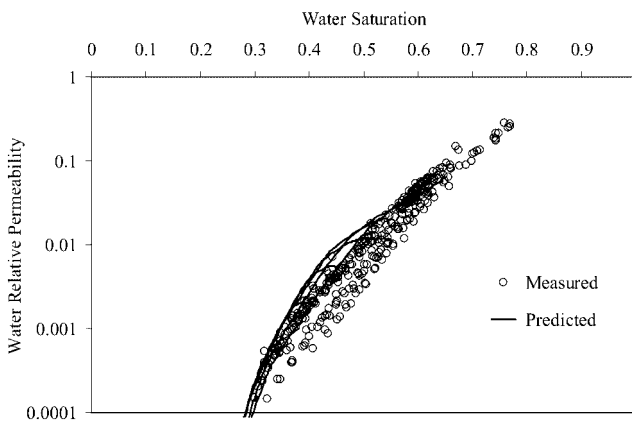


Figure 15.17 Comparison of measured and computed three-phase water relative permeabilities during gas injection (after Piri and Blunt, 2005b; courtesy of Dr. Mohammad Piri).

gas injection with the data of Oak (1990), while Figure 15.18 does the same, but for the gas phase. All the relevant features are predicted accurately by the model. As for the oil phase, the predicted RPs at high oil saturations are also accurate, but at low oil saturations where the flow is dominated by spreading layers, their pore network model systematically over predicts the oil RP. This is shown in Figure 15.19. The over prediction appears to be due to the overestimation of the oil layer conductance, hence implying that the simple representation of the conducting layers in a corner that they used fails to capture parts of the pore space where the layers are much less stable or conductive.

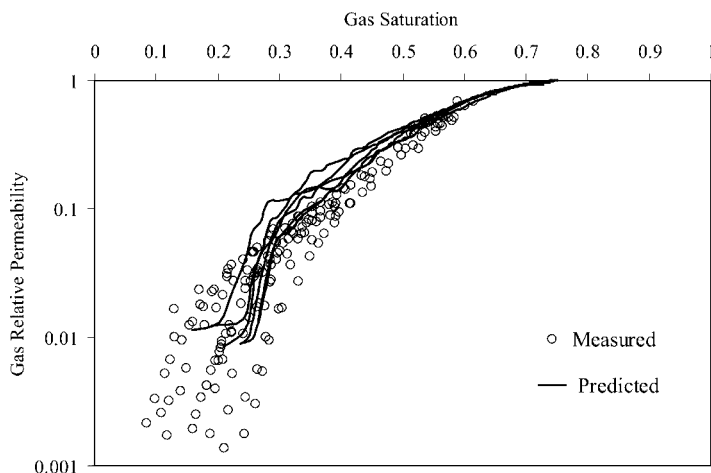


Figure 15.18 Comparison of measured and computed three-phase gas relative permeabilities during gas injection (after Piri and Blunt, 2005b; courtesy of Dr. Mohammad Piri).

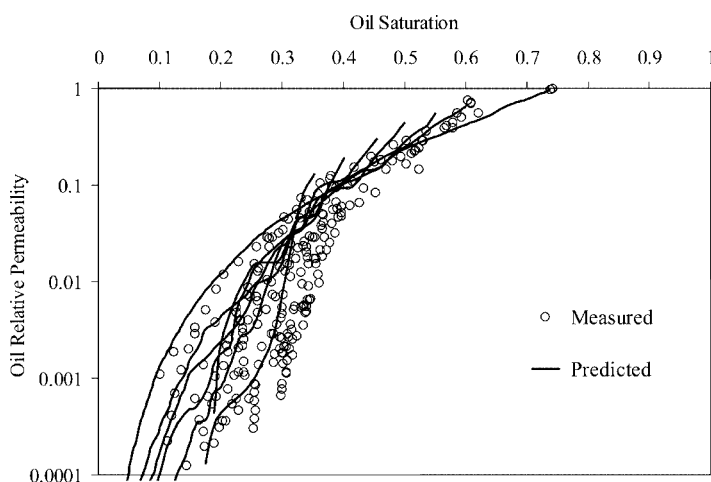


Figure 15.19 Comparison of measured and computed three-phase oil relative permeabilities during gas injection (after Piri and Blunt, 2005b; courtesy of Dr. Mohammad Piri).

Piri and Blunt also compared the RPs of a tertiary gas injection with those of a secondary gas injection with similar initial oil saturation. The oil RP for the tertiary process was nonzero and larger than that of the secondary gas injection at low oil saturations, because the double displacement caused an increase in the oil/water capillary pressure – in the tertiary gas injection – leading to thicker and more conductive oil layers. See also Fuller *et al.* (2006) for the important application to CO₂ sequestration, and Suicmez *et al.* (2007) for the WAG process as well as Suicmez *et al.* (2008).

15.15

Two-Phase Flow in Fractures and Fractured Porous Media

Similar to single-phase flow and dispersion, two-phase flow in fractures and fractured porous media has been studied for years. The phenomenon is important for enhanced oil and gas recovery, isolation of radioactive waste, exploitation of geothermal fields for generating electricity, and recovery of coalbed methane. The conventional approach has been based on the assumption that Darcy's law is applicable to both fluid phases. Moreover, it is usually assumed that the RP to each phase is equal to its saturation but, as described in Chapter 14, the assumption has been questioned, and more data are accumulating that indicate that the RPs in fractures may be more like those in unfractured porous media.

In the petroleum engineering literature, and to some extent, in the groundwater community, the double-porosity model that was described in Chapter 12 has been used for simulating two-phase flows in fractured porous media. However, given that two-phase flows crucially depend on the connectivity of the fluid phases, and that the double-porosity model completely ignores this important effect – as it assumes that the fracture network is well connected, while the porous matrix is disconnected – it is clear that the double-porosity model cannot be a useful for most cases.

As described in Chapters 6, 8 and 12, due to its rough internal surface, a single fracture can be mapped onto a equivalent porous medium and, hence, the application of pore network models to modeling of multiphase flow in a fracture is natural. Indeed, a few papers have attempted to use this approach. Haghighi (1994) and Haghighi *et al.* (1994) were probably the first who used micromodels to study imbibition and drainage in a fractured system. They also developed a network model to study the same phenomena using a model similar to the IP.

Glass *et al.* (1998) used a modified form of the invasion percolation to model quasi-static immiscible displacement in horizontal fractures. To estimate the invasion pressure, the effect of the contact angle, the local aperture, and the local in-plane interfacial curvature between the fluid phases was taken into account. A dimensionless parameter, the curvature number C_n that weighs the relative importance of the in-plane curvature and aperture-induced curvature, was introduced. As C_n was raised from zero, the invasion fronts made a transition from invasion-percolation type to smooth fronts. A somewhat similar model was also used by Amundsen *et al.* (1999).

A more refined model was developed by Hughes and Blunt (2001) for studying wetting in a single fracture. The fracture was represented by a square network of pore bodies and throats, and the effects of wetting layers, snap-off, and piston-like displacement that were described earlier were included in the model. The same type of model, but more refined in the manner that was developed by Piri and Blunt (2004, 2005a,b), was developed by Karpyn and Piri (2007) and Piri and Karpyn (2007) to compute the relative permeabilities and fluid occupancy statistics in a single fracture. The fracture, which had rough internal surfaces, was mapped with high resolution using X-ray microtomography. The relative per-

meabilities were computed, and piston-like displacement, cooperative pore filling, and snap-off were all included in the model. The simulation results agreed with the experimental observations of Karpyn *et al.* (2007). Bogdanov *et al.* (2003) studied two-phase flows in a fractured porous medium using the 3D model of discrete fracture networks that was developed by Adler, Thovert, and their collaborator and described in Chapters 8 and 12.

A novel model was proposed by Sahimi *et al.* (2010) who developed a new algorithm for generating models of fractured porous media. Similar to Bogdanov *et al.* (2003), they used discrete fracture modeling in which all the interactions and fluid flow in and between the fractures and within the matrix were modeled in a unified manner using the same computational grid. In their model, the geological model (GM) – the high resolution computational grid – of a fractured porous medium is first generated using square or cubic grid blocks. The GM is then upscaled using a method based on the multiresolution wavelet transformations (Mehrabi and Sahimi, 1997; Ebrahimi and Sahimi, 2004; Rasaei and Sahimi, 2008, 2009a,b). The upscaled grid contains a distribution of the square or cubic blocks of various sizes. A map of the blocks' centers is then used with an optimized Delauney triangulation method and an advancing-front technique in order to generate the final unstructured triangulated grid suitable for use in any general reservoir simulator with any number of fluid phases. The model also included an algorithm for generating fractures that, contrary to the previous methods, does not require modifying their paths due to the complexities that may arise in spatial distribution of the grid blocks. It also included an effective partitioning of the simulation domain that results in large savings in the computations times. The speed-up in the computations with the new upscaled unstructured grid is about three orders of magnitude over that for the initial GM.

The field of two-phase flow in fractures, fracture networks, and fractured porous media remains largely undeveloped. The classical double-porosity models have proven to be grossly inadequate in many cases and, therefore, much more research is needed.

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