PREFACE

FOR A COMPLEX TECHNOLOGY to reach a productive maturity, the disparate scientific disciplines underlying that technology must be gathered into a coherent whole. It is difficult to imagine a technology for which this is better exemplified than the geological disposal of radioactive wastes. The underlying scientific areas include surface chemistry (sorption–desorption, dissolution, ion exchange, corrosion), solution chemistry (hydrolysis, complexation, oxidation–reduction, precipitation), colloid chemistry, ceramics, metallurgy, hydrology, rock mechanics, and geology. These and other fields of study must be synthesized into a useful, practical whole through modelling and rational thought. Finally, the rationalized body of information and conclusions drawn must be substantiated by tests performed in the actual waste disposal environment.

This volume makes an important contribution to the information needed for disposal of wastes in geological media, demonstrating the advanced state of knowledge in many of the above fields of research. It represents a major part of what must be known before high-level radioactive waste disposal may become a reality.

Governmental agencies in the United States and other countries have sponsored a large amount of research on the behavior of radioactive wastes in various environmental settings. The overall objective of this research has been to protect the health and safety of the public by assessing the potential hazard of radionuclides in disposed wastes over periods of time when these radionuclides are significantly active. Making such an assessment requires an understanding of radionuclide distribution and inventory in or near the disposal site, and of the transport processes (chemical, physical, and biotic) that control the movement of radionuclides. The geochemistry of radionuclides in disposal environments is clearly one of the most important aspects of safety assessment because radionuclide release from the disposal site is controlled by complex chemical processes. The goal of this volume is to provide the reader with a single source of the most recent and significant findings of research on the geochemical behavior of disposed radioactive wastes.

Radioactive wastes of concern include wastes that result from operation of the nuclear fuel cycle (mining, fuel fabrication, reactor operation, spent fuel reprocessing, and waste storage), from nuclear weapons testing, and from medical and research activities. In recent years, the emphasis has been on predicting the behavior of disposed high-level wastes in deep geologic

repositories. The many chapters on high-level waste reflect this emphasis. However, the chemical behavior of the individual radionuclides described will apply to many types of waste in geologic environments.

The chapters of this volume are organized into sections that cover the chemical aspects that are important to understanding the behavior of disposed radioactive wastes. These aspects include radionuclide sorption and desorption, solubility of radionuclide compounds, chemical species of radionuclides in natural waters, hydrothermal geochemical reactions, measurements of radionuclide migration, solid state chemistry of wastes, and waste-form leaching behavior. The information in each of these sections is necessary to predict the transport of radionuclides from wastes via natural waters and thus to predict the safety of the disposed waste.

Radionuclide transport in natural waters is strongly dependent on sorption, desorption, dissolution, and precipitation processes. The first two sections discuss laboratory investigations of these processes. Descriptions of sorption and desorption behavior of important radionuclides under a wide range of environmental conditions are presented in the first section. Among the sorbents studied are basalt interbed solids, granites, clays, sediments, hydrous oxides, and pure minerals. Effects of redox conditions, groundwater composition and pH on sorption reactions are described.

Solubility constraints define the maximum concentrations of radionuclides at the point of release from the waste. In the second section, radionuclide solubilities in natural waters are reported as measured values and estimated values from thermodynamic data. In addition, information is given concerning the chemical species of radionuclides that could be present in natural waters.

If the heat generated from the waste by radioactive decay is great enough (as in the case of high-level waste disposed of in deep geologic repositories), hydrothermal reactions will occur between the groundwater, host rocks, and waste. The resulting alteration of these solids and groundwaters will affect the behavior of radionuclides in these systems. In the third section, the effects of these hydrothermal reactions are described.

Field measurements of radionuclide migration can be used to help substantiate laboratory measurements of sorption, solubility, and identification of important chemical species. The fourth section describes three field investigations that provide information on the effects of organics, colloids and environmental conditions (Eh, pH, and temperature) on radionuclide transport. The chemical species of radionuclides that are mobile under specific field conditions are identified.

Solid state chemistry of potentially important waste forms is covered in the fifth section. Solid state reactions can determine the oxidation state and physical and chemical stability of radionuclides in various host waste forms. This information can be used to evaluate the utility of crystalline materials as potential hosts for radioactive wastes. Groundwater leaching of radionuclides from waste forms is the first step in radionuclide transport from a disposal site. The release rate of radionuclides from the waste form is dependent on the waste form's leaching behavior. The sixth section describes the factors that affect the leaching behavior of several potential waste forms and radionuclides.

Finally, Mike McCormack, former Washington state Congressman, discusses the Federal legislation affecting nuclear waste disposal in the United States and the impact of several new laws passed by the Congress—the Nuclear Waste Policy Act of 1982 and the Low-Level Radioactive Waste Policy Act of 1980.

This volume covers ongoing research and, thus, leaves many questions unanswered and many problems unsolved. The geochemistry of disposed radioactive wastes involves many complex issues that will require years of additional research to resolve. High-priority problems include: integration of geochemical data with computer models of chemical interaction and transport, definition of environmental conditions that affect the behavior of radionuclides at specific disposal sites, evaluation of complex formation of dissolved radionuclides with inorganic and organic complexants, and determination of radionuclide solubilities in natural waters.

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