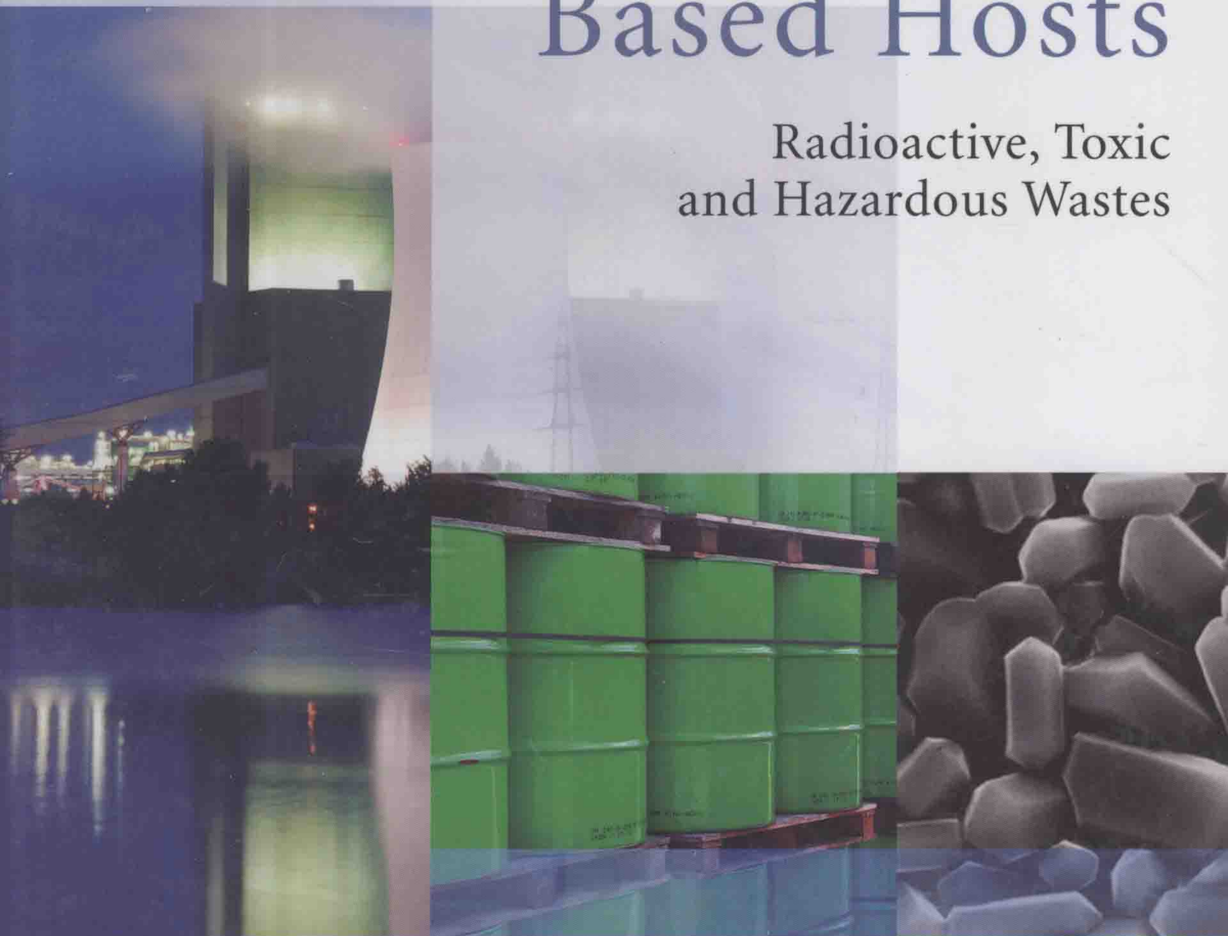
Ian W. Donald

Waste Immobilization in Glass and Ceramic Based Hosts

Radioactive, Toxic and Hazardous Wastes



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Preface

Over the last few years the problem of what to do with waste materials both radioactive and, more recently, nonradioactive, has become an increasingly important environmental and political issue.

In the case of radioactive wastes from reprocessed nuclear fuel and certain defence operations, vitrification has been the vanguard of past and current treatment options. More recently, the threat of global warming and the need for low carbon energy sources has brought nuclear issues back to the forefront of scientific, technical and public awareness, with the building of new nuclear power plants, coupled with the longer term likelihood of a future generation of advanced closed fuel cycle nuclear energy systems, the so-called Generation IV systems (GenIV). As worldwide demand for uranium increases with these new requirements, and stocks are depleted, fuel recycling, with the corresponding generation of various categories of radioactive wastes, will become an ever more important issue. Of serious concern at this time is the lack of specialists in areas relating to nuclear power and particularly nuclear waste, and this book provides an up-to-date reference source at this critical time, with the emphasis on waste immobilization. On the other hand, it is becoming increasingly essential from an environmental perspective, as regulated land-fill sites become full and/or more expensive, that more be done to address problems associated with the ever increasing volume of nonradioactive hazardous wastes, until recently treated with somewhat less respect than their radioactive counterparts. One solution to this problem is to turn hazardous wastes into vitrified or ceramic products where the hazardous elements are chemically immobilized and made passively safe, and it is in this area that certain practical lessons can be learnt from the nuclear industry with its long history and experience of waste treatment and vitrification technologies. An added advantage and incentive in the case of nonradioactive wastes is that, rather than generating a product that must be disposed of, treatment may actually turn hazardous wastes into useful products with practical applications.

This book brings together all aspects of waste immobilization, draws comparisons between the different types of wastes and treatments, and outlines where lessons learnt in the radioactive waste field can be of benefit in the treatment of nonradioactive wastes. These are areas very much of topical interest, rarely covered together despite the similarities that can be drawn. A wide range of topics is covered, beginning with introductory chapters which outline environmental aspects and provide information relating to generic sources and categories of wastes, potential disposal options, and where lessons can be learnt from nature and from certain incidents and accidents that have occurred from time to time. In the following chapters, immobilization of high level waste and special categories of intermediate level waste are covered in depth. Details of specific waste systems are provided, including the special categories of waste generated, for example, by the pyrochemical reprocessing of Pu metal, together with surplus materials not until recently considered as wastes, in addition to the more conventional wastes from spent fuel reprocessing and defence operations. Vitrification techniques, the types of glasses employed or proposed as wasteforms, glass-ceramic based hosts, glass-encapsulated ceramic wasteforms, and important ceramic phases are discussed. The characterization of radioactive wasteforms, a key issue with important implications for assessing their very long-term stability, is addressed. Nonradioactive hazardous and toxic wastes and wasteforms are subsequently reviewed in depth. These wastes are numerous and diverse in nature, ranging from municipal incinerator ashes to waste asbestos products, and currently create serious environmental challenges. The influence of microbial activity on wasteform stability, both radioactive and nonradioactive is also reviewed. This is an important topic which, to date, has only received limited attention but which could have serious implications for the long-term behaviour of wasteforms, particularly in repository and storage or disposal environments. The final chapter highlights comparisons between nonradioactive and hazardous waste immobilization and suggests where lessons learnt by the nuclear industries over many years may be usefully applied to the immobilization of nonradioactive hazardous waste systems. Finally, an outlook for the future is offered, with particular reference given to technological advances and the treatment of new generation wastestreams.

The book is particularly aimed at scientific and technical staff in the nuclear and waste management industries in addition to universities and research organizations active in these areas. It will also appeal to a wider audience with interests in glass or environmental issues and will be of benefit to anyone who requires background information on radioactive issues connected with nuclear energy or defence processes, or hazardous waste sources, properties and treatments. The provision of a comprehensive bibliography makes this book an up-to-date reference source in the areas of both radioactive and nonradioactive waste immobilization.

Ian Donald
September 2009

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List of Abbreviations

AEA	Atomic Energy Authority (UK)
AGR	Advanced Gas Cooled Reactor
ALARP	As Low As Reasonably Practicable
ALMR	Advanced Liquid Metal Reactor
ANL	Argonne National Laboratory (USA)
ANSTO	Australian Nuclear Science and Technology Organization
AVH	Atelier de Vitrification de La Hague (France)
AVM	Atelier de Vitrification de Marcoule (France)
AWE	Atomic Weapons Establishment (UK)
AWRE	Atomic Weapons Research Establishment (predecessor of AWE)
BFS	Blast Furnace Slag
BNFL	British Nuclear Fuels Ltd
BPM	Best Practicable Means
Bq	Becquerel
BWR	Boiling Water Reactor
CANDU	Canadian Deuterium Uranium reactor
CCT	Continuous Cooling Transformation curve
CEA	Commissariat à l'Energie Atomique (France)
Ci	Curie
CNWRA	Center for Nuclear Waste Regulatory Analyses (USA)
COGEMA	Compagnie Générales des Matières Nucléaires
CORALUS	Corrosion of Alpha-active glass in Underground Storage
CRNL	Chalk River Nuclear Laboratories (Ontario, Canada)
DEFRA	Department of the Environment, Food and Rural Affairs (UK)
DOE	Department of Energy (USA)
DoE	Department of the Environment (UK)
dpa	Displacements per atom
DWPF	Defence Waste Processing Facility (USA)
EAFD	Electric Arc Furnace Dust
EDS	Energy Dispersive Spectroscopy
EELS	Electron Energy Loss Spectroscopy
EPA	Environmental Protection Agency (USA)
EURATOM	European Atomic Energy Community
EXAFS	Extended X-ray Absorption Fine Structure
FDA	Food and Drug Administration (USA)
FINGAL	Fixation In Glass of Active Liquors

FP	Fission Product
FTIR	Fourier Transform Infra-Red (spectroscopy)
GenIV	Generation IV (nuclear energy systems)
GFR	Gas-cooled Fast Reactor
GMOD	Glass Material Oxidation and Dissolution system
GT-MHR	Gas Turbine–Modular Helium Reactor
HARVEST	Highly Active Residue Vitrification Experimental Studies (Harwell Vitrification Engineering Study)
HEPA	High Efficiency Particulate Air filter
HEU	Highly Enriched Uranium
HLLW	High Level Liquid Waste
HLW	High Level Waste
HLWC	High Level Waste Concentrate
HMI	Hahn Meitner Institute (Germany)
HRTEM	High Resolution Transmission Electron Microscopy
HSE	Health and Safety Executive (UK)
HWVP	Hanford Waste Vitrification Plant (USA)
IAEA	International Atomic Energy Agency
ICP-AES	Inductively Coupled Plasma Atomic Emission Spectroscopy
ICPP	Idaho Chemical Processing Plant (USA)
IFR	Integral Fast Reactor
ILW	Intermediate Level Waste
INE	Institut für Nukleare Entsorgungstechnik (Germany)
INEEL	Idaho National Engineering and Environmental Laboratory (USA)
ISV	*In-Situ* Vitrification
JAERI	Japan Atomic Energy Research Institute
LAW	Low Activity Waste
LD_{50}	Dose for an expected 50% death rate
LEU	Low Enriched Uranium
LILW	Low and Intermediate Level Waste
LLNL	Lawrence Livermore National Laboratory (USA)
MAS NMR	Magic Angle Spinning Nuclear Magnetic Resonance
MCC	Materials Characterization Center (at PNNL, USA)
MDS	Molecular Dynamics Simulation
MoD	Ministry of Defence (UK)
MOX	Mixed Oxide (fuel)
MSW	Municipal Solid Waste
MWe	Megawatts electric
NAS (CISAC)	National Academy of Sciences (Committee on International Security and Arms Control) (USA)
NBO	Non Bridging Oxygen
NDA	Nuclear Decommissioning Authority (UK)
NGNP	Next Generation Nuclear Plant
NGR	Nuclear Gamma Resonance
NII	Nuclear Installations Inspectorate (UK)
NIREX	Nuclear Industry Radioactive Waste Executive (UK)

NRC	Nuclear Regulatory Commission (USA)
ORNL	Oak Ridge National Laboratory (USA)
PAMELA	Pilot Anlage Mol zur Erzeungung Lagerfähiger Abfälle
PBMR	Pebble Bed Modular Reactor
PCM	Plutonium-Contaminated Material
PCT	Product Consistency Test
PIVER	Pilot Verre
PNC	Power Reactor and Nuclear Fuel Development Corporation (Japan)
PNNL	Pacific Northwest National Laboratory (USA)
PUF	Pressurized Unsaturated Flow (test)
PUREX	Plutonium and Uranium Extraction (refining process)
PWR	Pressurized Water Reactor
SEM	Scanning Electron Microscopy
SHS	Self-propagating High temperature Synthesis
SIMS	Secondary Ion Mass Spectrometry
SPFT	Single Pass Flow Through (test)
SRS/P	Savannah River Site/Plant (USA)
STEM	Scanning Transmission Electron Microscopy
Synroc	Synthetic Rock
TCLP	Toxicity Characteristic Leaching Procedure (test)
TCS	Toxicity Classification System
TEM	Transmission Electron Microscopy
THORP	Thermal Oxide Reprocessing Plant (UK)
Tof	Time-of-flight
TRISO	Tristructural Isotropic (fuel particles)
TRU	Transuranic (elements)
TTT	Time Temperature Transformation (curve)
USEPA	United States Environmental Protection Agency
VHTR	Very High Temperature Reactor
WAK	Wiederaufarbeitungsanlage Karlsrhue (Germany)
WASRD	Waste Acceptance Systems Requirement Document
WDS	Wave Dispersive Spectroscopy
WIP	Waste Immobilization Plant (Tarpur, India)
WIPP	Waste Isolation Pilot Plant (USA)
WNRE	Whiteshell Nuclear Research Establishment (Canada)
WVDP	West Valley Demonstration Project (USA)
XAS	X-ray Absorption Spectroscopy
XANES	X-ray Absorption Near Edge Structure
XRD	X-Ray Diffraction
XRF	X-Ray Fluorescence

Contents

1

Introduction

Throughout history, human society has generated huge quantities of waste materials through everyday living. This is particularly true of such periods as the industrial revolutions in Europe and elsewhere. In the past, the overall management of wastes has been poor, with a general disposition to dispose of these directly into the environment through dispersal, and with little thought given to the long term consequences of this action on the environment. Fortunately, this has not in general been the case for radioactive waste materials. After the discovery of radioactivity and radioactive materials in the late nineteenth century it soon became clear that these posed a special risk to humans and the environment. Consequently, radioactive wastes could not be treated in the same haphazard way as their nonradioactive counterparts but needed to be contained and excluded from the immediate environment. It is only relatively recently that serious efforts and a similar approach have been made in order to deal with nonradioactive toxic and hazardous waste materials through treatment, waste minimization, or recycling.

1.1 Categories of Waste and Waste Generation in the Modern World

Radioactive wastes are generated as a consequence of numerous processes and operations. These range from the reprocessing of spent nuclear fuel and plutonium production for weapon applications, to mining and refining of uranium ore, commercial research activities and use of isotopes, and medical, hospital and university activities. Unprocessed spent nuclear fuel itself has also been considered as a waste, although as discussed later this view is changing. Radioactive waste management practices vary worldwide but share the common interest of treating these wastes as highly hazardous materials from which the environment must be protected. Nonradioactive toxic and hazardous wastes are also generated by a host of industrial operations ranging from municipal incinerators to ferrous and nonferrous metal manufacture and processing, and these too are now attracting more serious attention.

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