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AN INTRODUCTION TO NUCLEAR WASTE IMMOBILISATION

SECOND EDITION

M.I. OJOVAN • W.E. LEE

An Introduction to Nuclear Waste Immobilisation

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Second Edition

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We dedicate this book to senior sisters Olga and Christina.

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Contents

Preface to the Second Edition **xiii**

1 Introduction to Immobilisation **1**
- 1.1 Introduction **1**
- 1.2 The Importance of Waste **1**
- 1.3 Radioactive Waste **2**
- 1.4 Recycling **4**
- 1.5 Waste Minimisation **4**
- 1.6 Processing and Immobilisation **5**
- 1.7 Time Frames **5**
- Bibliography **6**

2 Nuclear Decay **7**
- 2.1 Nuclear Matter **7**
- 2.2 Radioactive Decay **7**
- 2.3 Decay Law **8**
- 2.4 Radioactive Equilibrium **9**
- 2.5 Activity **11**
- 2.6 Alpha Decay **12**
- 2.7 Beta Decay **12**
- 2.8 Gamma Decay **14**
- 2.9 Spontaneous Fission **14**
- 2.10 Radionuclide Characteristics **15**
- Bibliography **19**

3 Contaminants and Hazards **21**
- 3.1 Elemental Abundance **21**
- 3.2 Migration and Redistribution **23**
- 3.3 Potential Hazard of Nuclear Waste **25**
- 3.4 Relative Hazards **27**
- 3.5 Importance of Wasteform: Real Hazard Concept **28**
- 3.6 Wasteform Durability and Hazard Diminishing **29**
- Bibliography **30**

4 Naturally Occurring Radionuclides **31**
- 4.1 NORM and TENORM **31**
- 4.2 Primordial Radionuclides **31**

4.3 Use of Primordial Radionuclides for Dating **33**
4.4 Natural Nuclear Reactors **33**
4.5 Cosmogenic Radionuclides **34**
4.6 Natural Radionuclides in Igneous Rocks **35**
4.7 Natural Radionuclides in Sedimentary Rocks and Soils **36**
4.8 Natural Radionuclides in Sea Water **37**
4.9 Radon Emissions **37**
4.10 Natural Radionuclides in the Human Body **39**
Bibliography **39**

5 Background Radiation **41**
5.1 Radiation is Natural **41**
5.2 Dose Units **41**
5.3 Biological Consequences of Irradiation **43**
5.4 Background Radiation **45**
Bibliography **47**

6 Nuclear Waste Regulations **49**
6.1 Regulatory Organisations **49**
6.2 Protection Philosophies **49**
6.3 Regulation of Radioactive Materials and Sources **51**
6.4 Exemption Criteria and Levels **51**
6.5 Clearance of Materials from Regulatory Control – Moderate Amounts **53**
6.6 Clearance of Materials from Regulatory Control – Bulk Amounts **53**
6.7 Double Standards **55**
6.8 Dose Limits **56**
6.9 Control of Radiation Hazards **56**
6.10 Nuclear Waste Classification **58**
6.11 IAEA Classification Scheme **59**
6.12 Examples of Waste Classification **61**
References **64**
Bibliography **64**

7 Principles of Nuclear Waste Management **65**
7.1 International Consensus **65**
7.2 Objective of Radioactive Waste Management **65**
7.3 Fundamental Principles **66**
7.4 Comments on the Fundamental Principles **67**
7.5 Fundamental Safety Principles **72**
7.6 Ethical Principles **72**
7.7 Joint Convention **73**
7.8 International Cooperation **74**
References **74**
Bibliography **74**

8 Nuclear Waste Types and Sources **75**
8.1 Sources of Nuclear Waste **75**
8.2 Front-End and Operational NFC Waste **78**
8.3 Back-End Open NFC Waste **81**
8.4 Back-End Closed NFC Waste **83**
8.5 Back-End NFC Decommissioning Waste **85**
8.6 Non-NFC Wastes **87**
8.7 Accidental Wastes **90**
8.8 Global Inventory **95**
References **96**
Bibliography **96**

9 Short-Lived Waste Radionuclides **99**
9.1 Introduction **99**
9.2 Tritium **99**
9.3 Cobalt-60 **101**
9.4 Strontium-90 **102**
9.5 Cesium-137 **104**
Bibliography **106**

10 Long-Lived Waste Radionuclides **107**
10.1 Introduction **107**
10.2 Carbon-14 **107**
10.3 Technetium-99 **109**
10.4 Iodine-129 **111**
10.5 Plutonium **112**
10.6 Neptunium-237 **114**
10.7 Nuclear Criticality **114**
References **115**
Bibliography **115**

11 Waste Processing Schemes **117**
11.1 Management Roadmap **117**
11.2 Waste Life Cycle **117**
11.3 Pre-disposal **119**
11.4 Disposal **121**
11.5 Categorisation for Processing **123**
11.6 Selection of Processing Technologies **125**
11.7 Wasteforms **129**
11.8 Waste Packages **133**
11.9 Processing of NORM waste **133**
References **134**
Bibliography **134**

12 Characterisation of Radioactive Waste **137**
12.1 Approaches to Waste Characterisation **137**
12.2 Characterisation of Radiation Fields **138**
12.3 Sampling and Characterisation of Surface Contamination **140**
12.4 Waste Characterisation Techniques **143**
12.5 Characterisation of Waste Packages and Wasteforms **152**
12.6 Characterisation of Environment and Personnel **156**
Bibliography **158**

13 Pre-treatment of Radioactive Wastes **159**
13.1 Pre-treatment Objectives **159**
13.2 Collection and Segregation **159**
13.3 Adjustment **161**
13.4 Size Reduction **161**
13.5 Packaging **163**
13.6 Decontamination **164**
Bibliography **170**

14 Treatment of Radioactive Wastes **171**
14.1 Treatment Objectives **171**
14.2 Treatment of Aqueous Wastes **171**
14.3 Treatment of Organic Liquid Wastes **184**
14.4 Treatment of Solid Wastes **185**
14.5 Treatment of Gaseous and Airborne Effluents **198**
14.6 Partitioning and Transmutation **202**
Reference **202**
Bibliography **202**

15 Immobilisation of Radioactive Waste in Cement **205**
15.1 Cementitious Wasteforms **205**
15.2 Hydraulic Cements **206**
15.3 Cement Hydration **208**
15.4 Phase Composition of Hydrated Cements **214**
15.5 Cementation of Radioactive Wastes **215**
15.6 Modified and Composite Cement Systems **216**
15.7 Alternative Cementitious Systems **219**
15.8 Cementation Technology **225**
15.9 Acceptance Criteria **230**
References **231**
Bibliography **232**

16 Immobilisation of Radioactive Waste in Bitumen **233**
16.1 Bituminisation **233**
16.2 Composition and Properties of Bitumen **233**
16.3 Bituminous Materials for Waste Immobilisation **234**
16.4 Waste Loading **235**

16.5 Bituminisation Technique 236
16.6 Acceptance Criteria 239
16.7 Bitumen Versus Cement 241
Bibliography 244

17 Immobilisation of Radioactive Waste in Glass 245
17.1 Glasses and the Vitreous State 245
17.2 Glasses for Nuclear Waste Immobilisation 247
17.3 Immobilisation Mechanisms 250
17.4 Borosilicate Glasses 251
17.5 Cations in Silicate Glasses 253
17.6 Degree of Polymerisation 254
17.7 Role of Boron Oxide 256
17.8 Role of Intermediates and Modifiers 257
17.9 Difficult Elements 258
17.10 Selection Rules for a Nuclear Wasteform Silicate Glass 258
17.11 Phosphate Glasses 260
17.12 Glass Composite Materials 262
17.13 Vitrification Technology 263
17.14 Development of Vitrification Technologies 267
17.15 Calcination Processes 272
17.16 Cold Crucible Melters 275
17.17 *In Situ* Vitrification 278
17.18 Radionuclide Volatility 280
17.19 Acceptance Criteria 281
References 282
Bibliography 282

18 New Immobilising Hosts and Technologies 283
18.1 New Approaches 283
18.2 Crystalline Wasteforms 285
18.3 Radiation Damage 288
18.4 Actinide-Hosting Ceramics 290
18.5 Polyphase Crystalline Wasteforms: Synroc 292
18.6 Polyphase Wasteforms: Glass−Crystalline Composites 294
18.7 New Technological Approaches 295
18.8 Metal Matrix Immobilisation 301
References 305
Bibliography 305

19 Transport and Storage of Radioactive Waste 307
19.1 Transportation 307
19.2 Storage 312
19.3 SNF Storage 317
19.4 Storage Inventory 318
Bibliography 318

20 Nuclear Waste Disposal **321**
20.1 Disposal/Storage Concepts **321**
20.2 Retention Times **321**
20.3 Multi-Barrier Concept **322**
20.4 Disposal/Storage Options **323**
20.5 Role of the EBS **327**
20.6 Importance of NGB **330**
20.7 Transport of Radionuclides **330**
20.8 Disposal Experience **333**
20.9 Acceptance Criteria **334**
Bibliography **335**

21 Safety and Performance Assessments **337**
21.1 Safety Case **337**
21.2 Safety Requirements **338**
21.3 Safety Assessment Report **339**
21.4 Safety Assessment Process **341**
21.5 Cementitious Materials Performance **343**
21.6 Bitumen Performance **347**
21.7 Glass Performance **349**
21.8 Glass Corrosion Mechanisms **349**
21.9 Glass Performance in Confined Conditions (Geological Repository) **354**
21.10 Radiation Effects **356**
21.11 Research Laboratories **359**
21.12 Conclusion **359**
References **361**
Bibliography **361**

Preface to the Second Edition

Since the publication of the first edition of this book in 2005 significant progress has been made in managing radioactive wastes in many countries. The decommissioning programme in UK is on course and the Swedish, Finnish and French high-level radioactive waste (HLW) repository projects move on apace. However, issues such as the accident at Fukushima in Japan (which will generate significant wastes during the clean-up over the next century), the continuing clean-up programme at Chernobyl in Ukraine and the impasse at Yucca Mountain in the US highlight both the international nature of waste management and the difficulties we all face in solving 'the waste problem'. While the technical challenges of dealing with radioactive waste, as described in this book, are demanding the particular difficulty of persuading communities to host repositories for HLW mean it will be many decades before the waste problem is fully addressed at a global level. In this edition we again focus on nuclear waste immobilisation but with significantly wider scope so that the actual book title should be *An introduction to nuclear waste processing*. We have not only updated the technological achievements but also added new chapters covering important topics such as nuclear waste characterisation, selection of optimal processing (including immobilisation) schemes, storage and transportation.

Although many of the radioactive substances currently used are of artificial origin, radioactivity is a natural phenomenon with many natural sources of radiation in the environment. Radiation and radioactive materials have many beneficial applications, ranging from power generation to industrial and agricultural irradiators and radiolabelled compounds in medicine and scientific research. Radioactive waste associated with these applications is generated, containing a range of radionuclide concentrations in a variety of physical and chemical forms. There are various methods for processing and then temporarily storing waste prior to disposal, and there are several alternative methods for its permanent safe disposal, ranging from near-surface to geological disposal. Wide differences in waste compositions may result in an equally wide variety of options for the management of the waste; therefore, a proper scheme of waste classification is required before any waste processing can be done. Many schemes have been developed to classify or categorise radioactive waste according to its physical, chemical and radiological properties. Such characterisation is important as the end points (e.g. storage and disposal) and conditioning methods (e.g. immobilisation and packaging in containers) depend on the level of radioactivity and radionuclide lifetime.

The key issue with any disposal option is safety, which is mainly achieved by concentration and containment involving the isolation of suitably conditioned radioactive waste in a disposal facility. Containment uses many barriers around the radioactive waste to restrict the release of radionuclides into the environment. Such an approach is key to waste storage and disposal; it is termed the *multi-barrier concept* and is often called '*matreshka*' after the popular Russian doll, which has inside of each larger doll a smaller one, so that the total number of dolls (barriers) is large. The confining barriers can be either natural or engineered, that is, obtained via processing. The accepted approach is to use more reliable barriers for more hazardous waste, including engineered barriers, which result from the radioactive waste treatment and conditioning processes.

1 Introduction to Immobilisation

1.1 Introduction

We are living in a naturally radioactive world. Our universe developed 13.8×10^9 years ago from a high-energy density singularity, the so-called Big Bang. It then expanded and was cooled by this expansion, the current 2.725-K background radiation being remnant radiation from the first seconds after the Big Bang. Everything we see around us is the substance of an exploded supernova star 4.6×10^9 years ago. At that moment all possible isotopes of all elements entered the composition of the supernova. The shortest-lived radionuclides decayed quickly, still in the plasma and gaseous phase as revealed by the presence of these radionuclides in the radiation spectrum of newer supernovae. Cooled gas condensed and formed solid dust particles which agglomerated to form the first universal bodies, which collided with each other to form the first protoplanets. Long-lived radionuclides, which became part of the composition of the protoplanets, gradually decayed, although longer-lived radionuclides such as ^{238}U, ^{235}U, ^{232}Th, and ^{40}K did not decay completely. These are left as a reminder of the natural radioactivity evolution over the 4.6 billion-year lifetime of the Earth.

Until the invention of the X-ray tube by Roentgen in 1885, natural radiation was the only ionising radiation in existence. In 1896 natural radioactivity was discovered by Becquerel and since then has been used for medical and research purposes. Since the first artificial radioactive materials were produced by the Curies in the 1930s, they have been utilised for society's benefit in science, medicine, industry and agriculture. However, using natural and artificial radioactivities leads to waste products, many of which contain significant levels of radionuclides.

1.2 The Importance of Waste

Waste has accompanied human society from pre-history to today and no doubt will accompany our future. Historically, we have been careless about managing the waste we produce. Disposal of waste into the surrounding habitat has to date been the usual practice with little concern for the environment. However, we now realise that waste has to be managed properly to preserve the planet for our children. To quote the Native American saying, 'We do not inherit the Earth from our ancestors; we borrow it from our children.' This realisation has been slow to come about, and

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not even the so-called developed countries have in place functioning, comprehensive policies for waste management.

Waste from use of radioactivity is, in many but not all cases, radioactive. Society has approached the management of radioactive waste differently from the management of other waste types. Rather than diluting and dispersing it into the environment, we have decided to contain and confine it. This is the first time in the history of human civilisation that such a decision has been taken consciously, as a matter of ethical principle; encouragingly, this prudent approach is now being extended to other wastes.

1.3 Radioactive Waste

Radioactive waste is defined as material that contains, or is contaminated with, radionuclides at concentrations or activities greater than clearance levels as established by individual countries' regulatory authorities, and for which no use is currently foreseen.

The higher the concentration of radionuclides above established levels the greater the hazard the waste possesses. The hazard of radioactive waste also depends on the nature of the radionuclides and, at the same concentration, different radionuclides have different levels of hazard. The definition of radioactive waste is purely for regulatory purposes. A waste with activity concentrations equal to, or less than, clearance levels is considered non-radioactive. From a physical viewpoint, however, it *is* radioactive – although the associated radiological hazards are negligible. Radioactive waste is in part waste like any other which is non-radioactive. However, radioactive waste may be accompanied by significant levels of ionising radiation; hence, it requires not only immobilisation to prevent radionuclides spreading around the biosphere, but also shielding and, in some cases, remote handling.

While most of the nuclear wastes from military and civil uses of radioactivity have been stored safely, in some cases, such as at Hanford in the USA, ill-defined, highly active sludges were stored in massive but leaky steel drums (Figure 1.1A); and it is only now that such sites are being cleaned up. At Hanford all the liquid contained in the drums has been removed and construction of massive facilities for waste immobilisation is underway. The US$12.2-billion clean-up project at Hanford is equivalent to building two nuclear power plants (NPPs) and the construction site (over 26 ha) includes facilities for pre-treatment, low activity waste vitrification and high activity waste vitrification, as well as an analytical laboratory (Figure 1.1B).

The financial cost of cleaning up such sites and others where accidental releases of radioactivity have occurred, such as in Chernobyl (Figure 1.2A) in Ukraine and Fukushima in Japan (Figure 1.2B), is enormous.

In the UK, the development and operation of the nuclear industry has left a legacy of waste that will cost >£100 billion to clean up. Worldwide, public

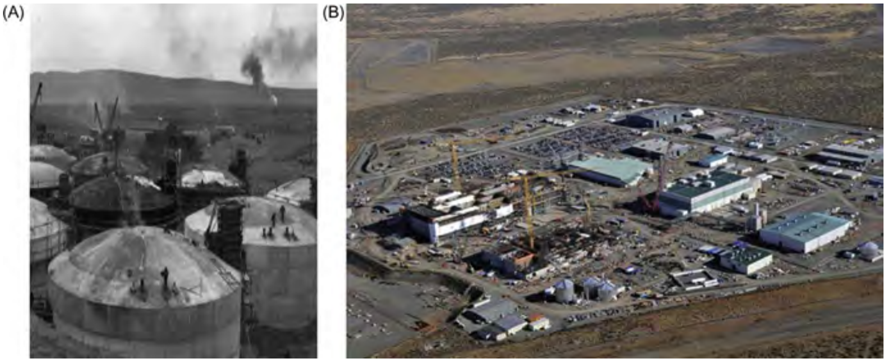

Figure 1.1 (A) Early picture of waste containers at Hanford. (B) A view of Waste Treatment & Immobilization Plant Project at Hanford (http://www.hanford.gov/page.cfm/WTP#).

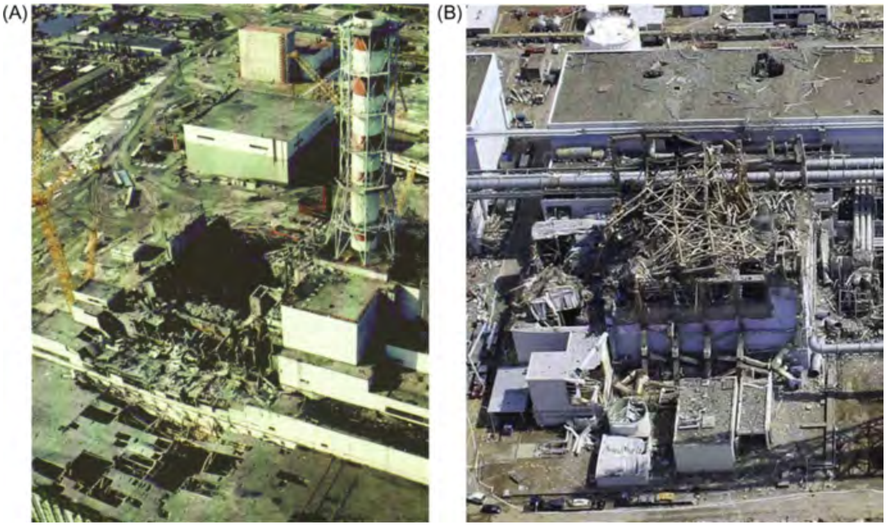

Figure 1.2 (A) The crater from the accident at Chernobyl, 1986. (B) Destroyed reactor block at Fukushima NPP, 2011.

perception of, and hence acceptance of, any future developments of new generating capacity involving NPPs will rely upon safe and efficient waste management. It is vital to demonstrate that these current waste problems can be resolved efficiently and safely and that new technology solutions can be applied directly to future wastes that may arise from new nuclear builds.

1.4 Recycling

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

One example of recycling in the nuclear industry is of spent nuclear fuel (SNF). A typical NPP generating 1 GW(e) produces approximately 30 t of SNF annually. The 435 nuclear power reactors currently operating worldwide produce about 10,500 t of spent fuel a year. During use, only about 5% of the uranium in the fuel is burnt, generating electricity but also forming transmutation products such as plutonium, which can be recycled, and minor actinides that may poison the fuel. After use, the fuel elements may be placed in storage facilities with a view to permanent disposal or be reprocessed to recycle their reusable U and Pu. Most of the radionuclides generated by the production of nuclear power remain confined within the sealed fuel elements. Currently, the world's spent fuel is often treated as waste and only a fraction of it is reprocessed in countries such as France and the UK. Recycling of fissile elements (U, Pu) from SNF, despite the complexity of such a process, results in a significant reduction of toxicity of the radioactive wastes.

Another potential example of recycling in the nuclear industry is of military-grade Pu, much of which is stockpiled in the USA, Russia, and the UK, a legacy of the cold war. It is possible to convert this material into a mixed U/Pu oxide (MOX) reactor fuel and programmes such as the USA/Russia PMDA (Pu Management Disposition Agreement) are now in place so that such material can be used to generate energy in a suitable nuclear reactor. A potential new development is 'inert matrix' fuel (IMF) which contains no U and in which Pu is the only fissionable component. This type of fuel would be optimised for burning of Pu, leaving a less dangerous spent fuel waste product.

It is likely that future generations will want access to our nuclear 'waste' and, as discussed by Burakov et al. (2010), there are many practical uses for radionuclides that will be developed by future generations.

1.5 Waste Minimisation

Waste minimisation is a process of reducing the amount and activity of waste materials to a level as low as reasonably achievable. Waste minimisation is now applied at all stages of nuclear processing from power plant design through operation to decommissioning. It consists of reducing waste generation as well as recycling, reuse, and treatment, with due consideration for both primary wastes from the original nuclear cycle and secondary wastes generated by reprocessing and clean-up operations.

Waste minimisation programmes were largely deployed in the 1970s and 1980s. The largest volume of radioactive waste from nuclear power production is low-level waste (LLW). Waste minimisation programmes have achieved a remarkable tenfold decrease of LLW generation, reducing LLW volumes to $\sim 100\ m^3$ annually per 1 GW(e). As a result of these waste minimisation programmes, the volume of waste from nuclear power generation has been further decreased and is incomparably smaller than that of fossil fuel generating the same amount of electricity. If all spent fuel was reprocessed, the high-level radioactive waste (HLW) from 1 year of global production of nuclear electricity could be accommodated by vitrification within the volume of a 10 m cube.

1.6 Processing and Immobilisation

Processing of radioactive waste includes any operation that changes its characteristics such as pre-treatment, treatment and conditioning. Conditioning may or may not include immobilisation.

Immobilisation reduces the potential for migration or dispersion of contaminants including radionuclides. The International Atomic Energy Agency (IAEA) defines immobilisation as the conversion of a waste into a wasteform by solidification, embedding or encapsulation. It facilitates handling, transportation, storage and disposal of radioactive wastes.

Conditioning means those operations that produce a waste package suitable for handling, transportation, storage and disposal. Conditioning may include, for example, the conversion of waste to a solid wasteform and enclosure of waste in containers. Conditioning thus is similar to immobilisation with the difference being in the scale. Conditioning is the engineering process dealing with large entities – packages.

1.7 Time Frames

Immobilisation is not a term used solely for radioactive waste. Many substances need a level of immobilisation or packaging during and after use. Immobilisation or packaging protects the substances contained and prevents access of the environment to them or their escape into the environment. However, in many applications, immobilisation is for relatively short time periods: from hours in medicine; days and months for food; and months and years to several tens of years for industrial chemicals. In the case of radioactive waste the immobilisation time required is extended to hundreds of years in the case of short-lived radionuclides, and thousands and hundreds of thousands of years for long-lived radionuclides. In addition, radioactive materials are continuously irradiating the immobilising medium, sometimes at significant levels, causing damage and structural changes. In the case of a reasonably short time, these changes can be understood and their impact on the wasteform taken into account. 'Wait and see' is not an option for time frames lasting hundreds of years. These two new features – extended times and irradiation – make immobilisation of radioactive wastes an issue which has no trivial solutions.

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