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Performance of engineered barrier materials in near surface disposal facilities for radioactive waste

Results of a co-ordinated research project

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FOREWORD

Member States' practices show that a large number of disposal operators use engineered barrier systems in their near surface disposal facilities for low and intermediate level radioactive waste (LILW). This is considered important for the protection of humans and the environment from the potential hazards associated with the disposal of radioactive waste, as well as to enhance public confidence in the safety of the disposal facility. The engineered barrier system can involve various components whose properties should be adequately identified in terms of their isolation behaviour and long term performance. These properties provide important information that serves as a basis for designing, conducting performance assessments and planning post-closure activities with respect to near surface disposal facilities.

A number of countries are planning or have already initiated research programmes on the performance of engineered barrier materials. The efforts are directed towards the establishment of an engineered barrier system which might be specific to planned or existing disposal facilities, as well as in support of the required barrier performance in an existing disposal facility. Investigators involved in such research programmes could benefit from sharing appropriate information. This situation led the IAEA to establish a Co-ordinated Research Project (CRP) on "Performance of Engineered Barrier Materials in Near Surface Disposal Facilities". The CRP was intended to promote research activities on the subject area in Member States, bring the participants together to share information, and contribute to advancing technologies for near surface disposal.

This report provides an overview of technical issues related to the near surface engineered barrier systems and summarises objectives, approaches and major findings of each individual research project that was carried out within the framework of the CRP. The report is expected to serve as a source of information for conducting research on and/or establishing an engineered barrier system for near surface disposal facility. The Scientific Secretary, K.W. Han of the Division of Nuclear Fuel Cycle and Waste Technology prepared the report in consultation with L. Nachmilner, Czech Republic, K. Philipose, Canada, R.G.G. Holmes, United Kingdom, and K. Kostelnik, USA. R. Dayal of the Division of Nuclear Fuel Cycle and Waste Technology finalized the report with the assistance of R. Little of the United Kingdom.

The IAEA wishes to express its thanks to all those who participated in this CRP and contributed to this publication.

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1. INTRODUCTION

1.1. BACKGROUND

The preferred option for disposal of short lived low and intermediate level radioactive wastes (LILW) is in near surface disposal facilities. The safety of the overall waste disposal system is determined by the performance of its individual components (waste form, waste container, engineered barriers and host environment) [1]. The backfill, barrier and cover materials are designed to perform an important role in preventing ingress of water, and in retarding radionuclide migration from the disposal facilities to the biosphere [2]. In the past, at some sites, lack of adequate attention in the selection of materials and in the engineering of the backfilling, sealing and covering of the trenches, vaults, ditches, etc., has resulted in the ingress of water and to the leaching of radionuclides from the disposed wastes [3].

It is important to select the appropriate backfill, barrier and cover materials and to engineer them in the most appropriate manner to backfill, seal and cover the disposal facilities. The extent to which this should be done depends on the types and properties of the wastes and site characteristics.

There is now four decades of operating experience in the design and operation of near surface disposal facilities [4, 5]. Various backfill barrier and cover materials have been used. Sand, gravel and different types of clays have been used as backfill and barrier materials to refill the disposal facilities. The cover of the disposal facility is usually a combination of materials such as sand, gravel, asphalt or concrete, and clay and soil conditioned for vegetation growth. The contouring of the cover is important. The general practice is to contour mounds to facilitate water run-off away from the disposed waste.

Operating experience in different countries has indicated successes and failures. It was recognised by the IAEA at the start of the 1990s that exchange and analysis of information on the performance of barrier and cover materials used in the past could provide useful insights to designers of new disposal facilities. Research on the performance of different materials with potential for use as engineered barriers in near surface disposal facilities was being undertaken in many countries, and it was recognized that the exchange of information on this work would be of mutual benefit. A co-ordinated research project (CRP) on this subject was established, considering that it would contribute to advancing technologies for near surface disposal and to improving the safety of future disposal facilities as well as promote the transfer of knowledge to developing countries.

The CRP was implemented from 1991 to 1995 during which three Research Co-ordination Meetings (RCMs) were held in 1992 (Keswick, United Kingdom), 1993 (Cordoba, Spain) and 1995 (Idaho Falls, United States of America). At the beginning, 13 research agreement contract holders from 13 countries participated. Italy could not continue its participation due to slowing down of its national programme on near surface disposal. On the other hand, an institute in China joined the CRP in the middle of the programme. France participated as an observer in the second RCM. Table 1 provides a list of participating institutions in the CRP and the topics that were investigated.

TABLE 1. INSTITUTIONS PARTICIPATING IN THE CRP

Country/organization (principal investigators)	Title of research	Area of application	Materials investigated	Performance measure investigated
Argentina/CNEA (J. Pahissa-Campa)	Study of Different Materials Available in Argentina to be Used as Barriers in Near Surface Disposal Facilities	Design	Backfill, Cap material	Sorption, Durability
Belarus/State University of Informatics&Radio-electronics (L. Lynkov)	Development of Technology for the Application of Barrier Composite Coatings, to the Surface of Low Level Radioactive Waste Disposal Facilities	Enhancement	Coatings-zirconium oxide	Sorption
Brazil/IPEN (A. Suarez/L.S. Endo)	Evaluation of Engineered Barrier Materials for Near Surface Disposal Facilities	Design	Concrete Cement	Diffusion, Durability
Canada/AECL (K.E. Philipose)	Assessment of the Long Term Behaviour of Repository Concrete	Assessment	Concrete	Durability
China/Inst. for Radiation Protection (G. Cunli/C. Anxi)	Performance Study on Engineering Barriers for Shallow Land Disposal of Low and Intermediate Level Waste	Design	Backfill	Sorption
China/CIAE (Z. Xin)	Study of Adsorption Formation of Nuclides on the Surface of Minerals	Backfill selection Assessment	Backfill	Sorption
Czech Republic/ Nuclear Research Institute (L. Nachmilner/ A. Vokal)	Optimisation of Engineered Barriers in Shallow Land Repositories	Assessment	Asphalt, Concrete, Bitumen	Durability
Hungary/ ETV-EROETERV (G.E. Szilagyine/ K. Berci)	Evaluation and Modelling of a Silty Clay Backfill and Cover Material for the Shallow Land Disposal of Waste Material	Assessment	Backfill/ cover (clay)	Sorption
India/BARC (N.K. Bansal/ P.K. Wattal)	Assessment of Multiple Barrier System Components for Waste Isolation in Near Surface Disposal Facilities	Enhancement	Grout, Wasteforms, Caps, Backfill	Leaching, Sorption
The Republic of Korea/ KAERI (H.H. Park/ K.S. Chun/W.J. Cho)	Selection and Performance Assessment of Engineered Barrier Materials	Design	Backfill	Diffusion, Permeability
Spain/ENRESA (P. Zuloaga)	Performance of Engineered Barriers Materials in Near Surface Disposal Facilities in Spain	Assessment	Concrete	Durability
UK/BNFL (R.G.G. Holmes)	Performance of Long Term Behaviour of Natural Clays and Cementitious Materials as a Barrier to Water Infiltration/Migration in a Near Surface Low Level Waste	Enhancement	Cement-clay wall, Clay caps	Permeability, Sorption
USA/Idaho National Engineering Laboratory (K. Kostelnik)	The US Department of Energy Buried Waste Integrated Demonstration	Remediation	Grout, Wasteforms, Backfill	Containment

1.2. OBJECTIVE

The primary objectives of the CRP were to:

- promote the sharing of experiences of the Member States in their application of engineered barrier materials for near surface disposal facilities;
- help enhance the use of engineered barriers by improving techniques and methods for selecting, planning and testing the performance of various types of barrier materials for near surface disposal facilities.

The objective of this publication is to provide an overview of technical issues related to the engineered barrier systems and a summary of the major findings of each individual research project that was carried out within the framework of the CRP. The report is expected to serve as a source of information for conducting research on and/or establishing an engineered barrier system for near surface disposal facility.

1.3. SCOPE

Research areas of the CRP focused on near surface engineered barrier systems and included mainly:

- application of engineered barriers;
- selection and characterisation of engineered barrier materials;
- performance of engineered barrier materials.

Also, exchange of technical information on relevant state-of-the-art technologies was undertaken through RCMs. Disposal to boreholes, several tens of meters below the Earth's surface, was considered to be beyond the scope of the CRP.

It should be noted at the outset that the output from the CRP is not comprehensive in terms of the barrier types, nor is it comprehensive in terms of timescales considered. The research was directed at barriers that were relevant to the needs of individual Member States who participated in the CRP.

1.4. STRUCTURE

This publication deals with a general overview of engineered barriers in near surface disposal facilities (Section 2), key technical information obtained within the CRP (Section 3) and overall conclusions and recommendations for future R&D activities (Section 4). Appendices presenting individual research accomplishments are also provided.

2. ENGINEERED BARRIERS FOR NEAR SURFACE DISPOSAL FACILITIES

2.1. NEAR SURFACE DISPOSAL CONCEPTS

Disposal options for LILW have been extensively reviewed by an IAEA group and the information recorded in a technical document [4]. The choice of disposal concept is driven by climate, waste form, geological and hydrogeological conditions, plus the assurance that the concept will meet the radiological performance requirements.

The various disposal options implemented by Member States have been described in Refs [4, 5] and can be summarised in a series of generic disposal concept categories. In all cases, the objective is to isolate waste from the human environment by controlling conditions within the disposal facility to limit, for example, water contact with waste, whilst ensuring the radioactivity is allowed to diminish either by natural decay or controlled dispersion. This latter option implies that the disposed waste will be controlled in terms of radionuclide content. The final feature that influences the selected disposal concept is the period of institutional control.

The generic concepts for near surface disposal can be described as follows.

- **Covered trench**: This is the oldest and simplest of the disposal concepts that consists of placing waste into excavated trenches and covering the filled trenches with soil. Typical of this concept is the original trench disposal system at the Drigg disposal facility in the UK [6]. Disposal sites using this concept frequently have retrofitted engineered barriers. The generic type is shown in Figure 1(a).
- **Closed vault**: This consists of a concrete vault into which is placed packaged and/or treated waste. The voidage may be backfilled and the structure closed with concrete slabs, which may be sealed by, for example, asphalt. The whole structure is then protected by an earthen cap (Figure 1(b)). Examples of this disposal concept may be found at the Centre de l'Aube in France [7], El Cabril in Spain [8] and Rokkasho-mura in Japan [9].
- **Domed vault**: This concept is best typified by the IRUS disposal facility in Canada [10] (see Figure 1(c)), in which infiltration is controlled by placing waste in a dry permeable layer and covering the waste with an impermeable concrete roof that is subsequently protected by an earthen cap.
- **Open vault**: In this concept, a low permeability cap is placed over the filled vault without emplacement of a concrete slab. Waste is however pre-treated to minimize voidage. The cap is designed to accommodate some settlement. This concept is used at the Drigg Site in the UK for the new vault disposal concept [6] and is shown in Figure 1(d).

In addition to the options involving trenches and vaults, near surface disposal facilities can include caverns tens of meters deep below the Earth's surface. The Republic of Korea conceptual disposal facility discussed in Appendix 10 is an example that was considered during the CRP. More generally, a range of cavern type, disposal facilities have been used and proposed for disposal of LILW, typically:

- specially excavated caverns;
- disused mines;
- natural cavities.

In the cavern type disposal facility, the surrounding host rock serves as the main barrier, e.g. against:

- external factors (e.g. erosion and human intrusion);
- radionuclide release;
- water ingress.

Other examples of such disposal systems are found at Asse and Morsleben in Germany, Forsmark in Sweden, and are described in Ref. [5].

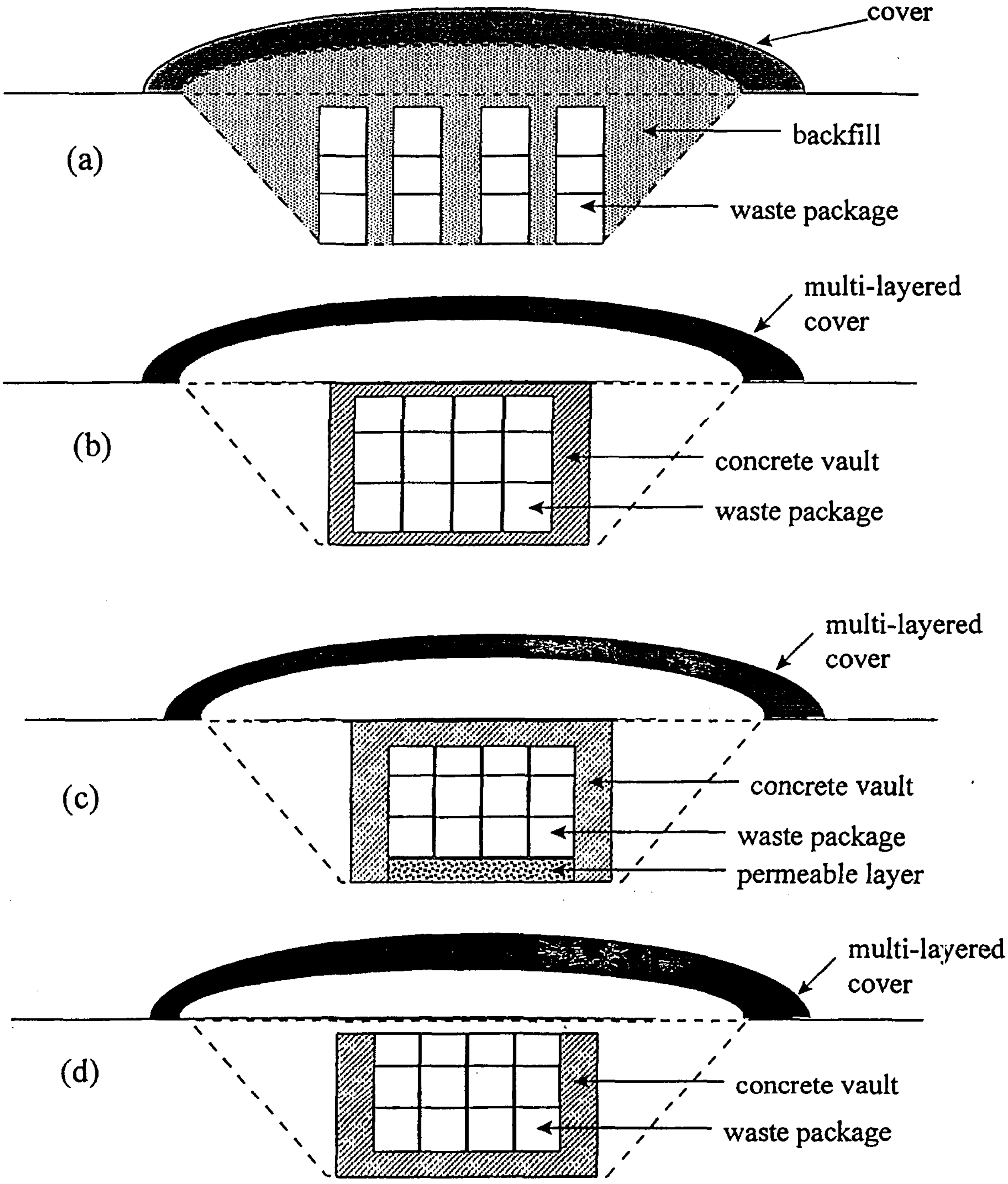

FIG. 1. Examples of near surface disposal facility concept (a: trench disposal, b: closed vault, c: domed vault, d: open vault).

2.2. FUNCTION AND NATURE OF ENGINEERED BARRIERS

There are a number of pathways by which radionuclides may migrate or be brought into contact with humans, including:

- infiltration of surface water;
- groundwater intrusion;
- subsequent migration of contaminated water (leachate);
- inadvertent intrusion;
- escape of radioactive gas.

Engineered barriers can be used as physical and/or chemical obstructions to prevent or delay the movement (e.g. migration) of radionuclides via these pathways. They are an integral part of the disposal facility and their incorporation is best achieved early in the design process. However, as additional information is obtained concerning waste characteristics, disposal site characteristics, indigenous material availability and disposal system performance, it should be incorporated into the disposal facility management plan as well as the engineered barriers' design to enhance the overall system performance. An example of this optimisation was presented by the UK participant (Appendix 12). At the Drigg site, it was envisaged that the refinement of BNFL's Engineered Barrier Model by means of the incorporation of performance data and design options would support future development as well as eventual closure of this site.

Engineered barriers can be used to limit the impact of the above pathways for a particular site as part of:

- the design of a new concept;
- the improvement of an operating site;
- the remediation of an existing or closed site.

The barriers represent an important component of the disposal facility safety from the operational phase, through the period of institutional control, and ultimately to the possible free release of the site.

The radiological performance assessment of the disposal facility can provide a key input into the choice of barrier. Crucial to the barrier choice is the timescale over which the barrier is assumed to function. This can be hundreds of years.

Barriers can provide either partial containment or complete isolation of waste, or both. Their efficacy depends on the interactions of natural and engineered barriers which, due to the complexity of interactions that can occur, may require some modelling or predictive capability to be fully understood and to ensure that the most effective barrier combinations are employed. The barriers can be described in terms of their function, for example, a cut off wall is used to limit horizontal migration of groundwater. Table 2 describes the barrier types that might be used to meet various functional requirements. In addition, Table 2 shows key parameters that are important in selecting a barrier.

TABLE 2. ELEMENTS OF ENGINEERED BARRIERS

Type	Function	Materials	Key parameters
Wasteform & container	– mechanical strength – limit water ingress – retain radionuclides	– cement grout – bitumen – polymers – concrete container – metallic container	– strength (compressive) – permeability – leach rate – time to failure/failure rate
Backfill	– void filling – limit water infiltration – sorption of radionuclides – precipitation of radionuclides – gas control – allow retrieval	– clays (natural & imported) – cement & clays – cement grout – clay mixtures (with cements, soils, rock etc.) – low strength infills	– sorptive capacity – permeability – porosity – mechanical properties
Structural material & liners	– physical stability – containment barrier	– concrete – reinforced concrete – clay – asphaltic or organic membranes – steel sheeting	– permeability – compressive strength – shear strength – thickness – time to failure/failure rate
Drains	– control leachate – enable monitoring	– gravel/sand – ceramic & cement	– permeability – rate of blockage
Cap	– limit water infiltration – control gas release – intrusion barrier (bio & human) – erosion barrier	– clays – asphalt or polymeric membranes – sand/soil – gravel/cobbles – geotextiles – concrete slabs – gravel/cobbles – vegetation	– permeability – water shedding capacity – plasticity – time to failure/failure rate

2.2.1. Wasteform and container

The wasteform and container are often selected to benefit other stages of waste management, e.g. storage and transport, as well as disposal. However, they can provide a barrier to radionuclide migration and provide some mechanical strength. The radionuclide barrier function can be provided by low permeability and/or by high sorption capacity. Strength (compressive) and leachabililty are important waste form properties.

2.2.2. Backfill

Backfills [11] are used for a number of purposes: void filling to avoid excessive settlement, limitation of water infiltration, sorption of radionuclides, precipitation of radionuclides, gas control and, if necessary to facilitate waste retrieval. Typical materials used, either singly or as admixtures, include clays, cement grout, rock, soil, etc. The key properties of backfill are: K_d to establish sorption capacity [12], resulting groundwater chemistry to aid precipitation of radionuclides, e.g. actinides or ^{14}C, permeability to control ingress of water and allow release of gases, and finally mechanical properties.

TABLE 3. MEMBER STATES' EXPERIENCE IN ENGINEERED BARRIERS

		SHORT TERM		LONG TERM		PREDICTIVE MODEL
FUNCTION	MATERIAL	PHYSICO-CHEMICAL PROPERTIES	DURABILITY	PHYSICO-CHEMICAL PROPERTIES	DURABILITY	
WASTE FORM	CEMENT	India, UK, Spain	Spain, UK	Spain, UK	Spain, UK	Spain
	BITUMEN					
	OTHERS	Belarus	Belarus	Belarus	Belarus	
BACKFILL	CLAY SOIL	China, Rep. of Korea, India, Argentina, UK, Hungary	UK	UK	UK	Korea, Rep. of Argentina
	CEMENT	India, Canada, UK (France), Hungary, Brazil, Czech Republic	UK, Canada (France), Brazil	UK, Canada, (France), Brazil	UK, Canada, (France), Brazil	Canada, (France)
	OTHERS	Czech Republic	Czech Republic	UK (France)	UK (France)	
WALLS	CEMENT	Spain, Canada UK (France)	Spain, Canada, UK (France)	Spain, Canada UK (France)	Spain, Canada, UK (France)	
	CLAY	UK	UK	UK	UK	
DRAINS			Spain	UK	UK	UK
CAPS	CLAY SOIL	Spain, UK, Hungary	Spain, UK	Spain, UK,	Spain, UK,	Spain, UK
	CEMENT	Czech Republic, Spain, UK	Czech Republic, Spain	Czech Republic, Spain, UK	Czech Republic, Spain	Spain
	POLYMER MEMBRANE	Czech Republic	Czech Republic	Czech Republic	Czech Republic	

2.2.3. Structural materials and liners

Disposal facility structure provides physical stability and containment barrier [13–16]. Structural materials and liners used for disposal facility structure include concrete, reinforced concrete, clay and asphaltic or organic membranes. Key properties that characterize the performance of disposal facility structure are permeability, compressive strength, and shear strength.

Cut-off walls [17] can be used to: limit horizontal migration of groundwater into or out of the disposal facility; provide structural integrity to a disposal facility during its operational phase; and, in some cases, assist in sorbing radionuclides to delay their release. Materials used as cut off walls are concrete curtain walls (also an integral part of a vault design), secant pile walls, cement-clay cut-off walls and steel sheeting. The key properties are effective permeability (i.e. the emplaced properties) and service life.

2.2.4. Drains

Drains are designed for water management, particularly during the disposal facility's institutional phase. Typical drainage systems are either combinations of clay and gravel blankets or conventional ceramic or concrete drains. The key parameter is volumetric flow.

2.2.5. Cap

The purpose of a cap is to facilitate water run-off, limit infiltration of water, provide gas control, and serve as an intrusion barrier and an erosion barrier [18]. Materials used in cap systems include clays, soil/sand, gravel and cobbles, geotextiles, concrete slabs, asphalt and polymeric membranes. The key parameters in selecting the cap design and its components include vertical permeability, water shedding capacity, plasticity, time to failure, etc.

2.3. EXPERIENCE OF MEMBER STATES

The participants in the CRP and other Members States of the IAEA have a wide range of expertise in the development and performance of engineered barriers associated with their research projects. These Member States represent a source of expertise that is summarized in Table 3.

3. CO-ORDINATED RESEARCH PROJECT CONTRIBUTIONS

As noted in Section 1.3, three research areas were considered in the CRP:

- application of engineered barriers;
- selection and characterisation of engineered barrier materials;
- performance of engineered barrier materials.

These are discussed sequentially in the following sub-sections.

3.1. APPLICATIONS OF ENGINEERED BARRIERS

Three applications of engineered barriers have been presented in this CRP. These applications include:

- engineered barriers included as part of near surface disposal facility designs;
- engineered barriers implemented as enhancements to operational near surface disposal facilities; and
- engineered barriers applied as remediation and site restoration actions to closed near surface disposal facilities.

3.1.1. Engineered barriers included as part of near surface disposal facility designs

3.1.1.1. Argentina

The Argentinian research presented was an example of engineered barriers being included in the design of new near surface disposal facilities. Argentina has in operation two nuclear power plants, the 380 MW(e) pressure vessel (PHWR) Atucha I and the 640 MW(e) pressure tube type Embalse. The Comisión Nacional de Energía Atómica (CNEA) has the responsibility to manage the country's entire nuclear fuel cycle. In 1986, CNEA established their waste management programme to deal with the treatment, conditioning, intermediate storage, transport and final disposal of low, medium, and high level radioactive waste, and to develop and implement all the necessary mechanisms to attain these objectives in compliance with Argentina's regulatory requirements. In support of this programme, Argentina has been conducting research on the behaviour and properties of indigenous clays for use as backfill components, coverings, and disposal beds.

3.1.1.2. Republic of Korea

Another example of engineered barriers being incorporated into the designs for near surface disposal facilities is a disposal facility for LILW in the Republic of Korea (Appendix 10). This below ground facility includes three types of caverns and engineered barriers for waste disposal depending on the waste activity. These include: the trash waste cavern; the solidified waste cavern; and the intermediate-level waste cavern for spent resins, spent filters, and high-activity trash wastes. The backfill materials have been a major component of the Korean disposal design. Compacted clay and crushed granite mixtures with various clay contents have been under investigation (Appendix 10). The radionuclide diffusion coefficients and their associated hydraulic conductivities have been investigated.

3.1.2. Engineered barriers implemented as enhancements to operational near surface disposal facilities

3.1.2.1. India

Solid and conditioned LILW is currently being disposed of at near surface disposal facilities in India. The safety assurance associated with these facilities is achieved through multi-barrier engineered structures. These barriers involve the primary barrier (i.e. waste form), and additional barriers such as backfill material, engineered structures and finally the hydrogeological features of the disposal site. In India, all disposal facilities are located at the nuclear power plants. Therefore, this situation establishes the disposal site's hydrogeologic conditions. The waste forms, backfill materials, and other engineered barriers are then evaluated for optimisation and are incorporated into the site design and operations.

Several areas of research have been presented (see for example Appendix 9). These include results on various backfill materials such as clay (including vermiculite), cement grout, and crushed rock. The objective is to improve waste form stability by filling void spaces. Radionuclide migration potential is also reduced by increasing sorption potential.

In addition to backfill materials, the implementation of other engineered barriers at India's disposal facilities include the use of various high-integrity containers. These containers serve as impervious barriers against the inflow of water as well as prevent migration of radionuclides from the disposal facility to the environment.

3.1.2.2. United Kingdom

A second example where engineered barriers are being implemented at operational disposal sites can be found at the Drigg site in the UK (Appendix 12). The Drigg Engineered Barrier Programme evolved over the period of the CRP. This has included research on cut-off walls, cap material and performance, and site/facility monitoring and modelling. Data are obtained for the model not only from in house studies but also from the extensive literature available for barrier and geologic materials.

During the CRP engineering work on the Drigg cut-off wall was ongoing. The objective of this work was to demonstrate the performance of a cement bentonite wall in controlling groundwater intrusion. The introduction of this cut-off wall has been shown to reduce the groundwater flow rates within the Drigg site as well as reduce the trace tritium leakage away from the Drigg trenches.

The Drigg vault monitoring programme continued throughout the duration of the CRP. This monitoring programme consisted of instruments placed in a series of locations under the disposal vault both in the gravel under blanket and in the underlying clay layer, which consists of both indigenous clays and a bentonite preparation. The final tests on the material ex-situ were reported (Appendix 12). These tests showed no catastrophic degradation of the underlying clay layer.

Work has been completed on two demonstration rigs at Drigg. The first of these, Rig 1, was used to investigate cap infiltration and lateral flow, the second, Rig 2, was used to investigate the effect of settlement on cap performance. Both of these studies demonstrated the importance of understanding the cap's behaviour and the performance requirements for both the site and the engineered barriers (Appendix 12).

3.1.2.3. Belarus

Additional research has been presented on innovative composite coatings (Appendix 1). These barrier coatings have been investigated for application to contaminated surfaces. Such coatings which resist degradation could extend the anticipated performance of a disposal facility.

3.1.3. Engineered barriers applied as remediation and site restoration actions to near surface disposal facilities

In cases where previously disposed of waste has or is likely to result in future contamination, it is desirable to minimise and or eliminate this potential migration. In this situation, means of encapsulating the disposed waste with additional engineered barriers is advantageous. Underlying floors, vertical walls, and caps can all be added to enhance the performance of a disposal facility.

3.1.3.1. United States of America

The United States Department of Energy (DOE) has initiated a comprehensive research, development, testing, and evaluation programme to provide innovative technology systems to remediate and achieve site restoration of closed near surface disposal facilities (Appendix 13). These innovative technology systems incorporate several aspects of engineered barriers.

As reported in Appendix 13, significant remediation challenges are presented by the buried hazardous and transuranic-contaminated (i.e. mixed) waste. An integrated systems engineering approach has been supported which has resulted in the development of several innovative remediation systems. These engineered systems include: the In Situ Stabilization and Containment System, and the Selective Retrieval System.

The In Situ Stabilization System presented involves the encapsulation of previously disposed waste (Appendix 13). This system showed that improved stability to the waste form was achievable if the waste cell was solidified. This approach also reduces the hydraulic conductivity of the disposal cell thereby reducing both surface and groundwater intrusion. Researchers also (a) investigated the spread of plutonium surrogates during the injection and removal operation, (b) determined the effectiveness, durability and efficiency of grouting a soil/waste matrix, (c) assessed the retrievability of a grouted soil/waste matrix, (d) investigated grout emplacement techniques for high clay-content, low permeability soils, and (e) investigated improved waste forms such as vitrified, cementitious and polymer grouts.

Additional technology systems that provide environmental and worker protection include equipment for remote selective retrieval (Appendix 13). This equipment removes the workers from the hazardous and radioactive contaminants while permitting segregation of various waste types for conditioning, repackaging and improved management. This approach adds an additional degree of safety to the waste management and remediation operations.

3.2. MATERIALS FOR ENGINEERED BARRIERS

The CRP addressed a number of the barrier types and their key parameters. In addition, a description of integrated barrier systems for the participating nations was provided as a component of the CRP. This section describes, by way of example, the results of the CRP against the key parameters for the barrier materials. The information presented below describes the properties of the material in its fresh and 'as placed' state. Of equal and perhaps greater importance is the evolution of these properties with time. This information is presented in Section 3.3 (see below) under Performance of Engineered Barriers.

It should be noted that the information on materials is not comprehensive (i.e. not all barrier materials were covered in the CRP). In addition, data are often specific to a particular material, formulation and method of production. The various engineered barriers and the materials used therein are described in the following text.

3.2.1. Wasteform

The efficacy of the waste form is established based on leach tests. A cement waste form favoured in India showed a leach rate lying between 2×10^{-4} and 3.0×10^{-6} g/cm^{2}day, whilst a polymer modified cement was tested by the Czech Republic (Appendix 7). These data are very dependent on the waste form composition and test conditions. In many safety cases, no benefit is claimed for the integrity of the waste form, other than its sorption capacity.

3.2.2. Backfill

Within the CRP, a range of backfill or potential backfill materials have been investigated. One of the most comprehensive studies is of Younil bentonite from the Republic of Korea admixed with crushed Daeduk, Taejon granite. Hydraulic conductivity (permeability) varies with clay concentration from ca. 1×10^{-6} m/s at 20% clay to 1×10^{-11} m/s at 100% clay. Effective diffusion coefficients for ^{125}I (a conservative tracer) and ^{90}Sr, are in the range 3.8×10^{-11} m^2/s to 7.12×10^{-11} m^2/s and 1.21×10^{-12} m^2/s to 1.43×10^{-12} m^2/s respectively. This indicates some retention of Sr by the clay.

In a related study on sodium bentonite, Cs, Tc, Sr and I were investigated in terms of their K_d values. The values of both Cs and Sr were high (up to 2×10^0 m^3/kg and 5×10^{-1} m^3/kg respectively) with the anionic species of Tc and I exhibiting a very low K_d (Appendix 6). Bentonites and Vermiculites indigenous to India were also tested for Cs, Sr and Co sorption, with high sorption capacities noted in all cases (Appendix 9).

ENRESA have reported on a cementitious backfill that shows a rapidly evolving porewater chemistry with high pH (pH 13.9–12.8) from NaOH and KOH in the grout (Appendix 11). At this pH, actinides will precipitate onto the grout surface.

3.2.3. Cut-off walls

These fall into two categories:

- concrete curtain walls that also form a part of the vault structure. These materials have a bulk permeability of ca. 1×10^{-13} m/s. These walls rarely fail through bulk permeability, but usually by cracking or poor sealing of the wall to the underlying low permeability layer.
- cement-bentonite cut-off wall such as that used at Drigg in the UK. This has been used to protect a portion of the site from inflow of groundwater. The mixture bentonite, Ordinary Portland Cement and granulated blast furnace slag has a bulk permeability of 1×10^{-9} m/s after a year of curing (Ref. Trivedi and Holmes). The wall itself has a permeability that is dependent on keying into the underlying layer of low porosity clay and the engineering of the wall. Tests on a cell set into the wall suggest an effective permeability of 1×10^{-8} m/s which is adequate, compared to the permeability of the local geology, to reduce water flow into the disposal facility.

According to results presented at the RCM in Keswick (UK), trials on the bentonite cut-off wall material to investigate the ability of the wall to sorb radionuclides showed that the Sr K_d was 0.046 m^3/kg, Cs 0.68 m^3/kg, Pu 6.53 m^3/kg, U 0.45 m^3/kg. Similar tests on concrete representative of a curtain wall gave K_d values as follows: Sr 0.016 m^3/kg; Cs 0.071 m^3/kg; Pu 14.18 m^3/kg; and U 16.08 m^3/kg.

3.2.4. Drains

The use of drainage systems is a feature of many concepts. For example, ceramic drains are used at El Cabril, whilst the Drigg concept relies on a gravel blanket over an engineered clay horizon (bentonite and crushed limestone). Trials were carried out at Drigg to investigate the possibility of attack of the engineered clay by an alkaline plume (Section 3.3.3). Modelling was also used to predict the behaviour of the drainage blanket, flow, silting and deposition of $CaCO_3$. These trials also gave comfort of a prolonged drain life.

3.2.5. Cap

Caps fall into two main types. The first is a cap that includes a rigid concrete slab that encloses the emplaced waste whilst supporting the cap structure, and the second is an unsupported earthen cap over waste which allows for some settlement of the waste.

3.2.5.1. Concrete slabs

The characteristic strength of the concrete slabs has been specified for the El Cabril site as 350 kg/cm^2 (Appendix 11). The permeability of typical structural concrete was reported as up to 1×10^{-13} m/s at the RCM in Keswick (UK). The effective permeability of a concrete slab is, however, likely to be substantially higher than this due to leakage around the emplaced slab. In a separate study, reported by the participants from the Czech Republic, the slab material permeability has been expressed in terms of porosity with a value of 37.7 m^3/kg.

3.2.5.2. Low permeability clays

The most common source of the large volumes of material required for extensive capping of disposal facilities is indigenous clays; these may be both readily available and inexpensive. The specification for the clays is usually set by the acceptable limit for the infiltration rate through the cap which is a function of the climate and can be estimated as part of the performance assessment of the disposal facility. In the CRP a wide range of materials have been used or investigated. The permeability has been measured as 6.4×10^{-8} to 4.1×10^{-10} m/s for Drigg Clay (reported by R Holmes) and 3.26×10^{-11} m/s for natural bentonite found in China (reported by Z Xin). Clays in conjunction with a more permeable layer will, in an unsupported cap, require an ability to settle. In trials carried out in the UK tests on a multilayer cap, with Drigg Clay and a layer of 1.7% soil, 10% sand with 20% kaolin, settlement of 500 mm could be tolerated without loss of performance. At a pitch equivalent to that of the proposed Drigg cap, this arrangement was able to shed the local rainfall (ca. 0.75 m/year) experienced by the cap without significant vertical infiltration.

3.2.6. Other barriers

The above barriers are largely associated with design of new facilities or improvement of existing operations. They may also be used remedially on old or closed facilities. It may however be necessary to employ innovative remedial solutions such as in-situ grouting or vitrification, emplacing polymer membranes (Appendix 13) and formation of tie-down coatings on soil particles using reactive organometallics (Appendix 1) to reduce leachability of contaminants.

3.3. PERFORMANCE OF ENGINEERED BARRIERS

Since the disposal safety concept often requires long term (several hundred years) performance of engineered barriers, durability of materials is an important aspect to meet the performance requirements of the near surface disposal facilities. As described in Section 2, many types of materials are used in engineered barrier applications. This sub-section describes the research work by the CRP participants to establish the required performance of materials for their LILW disposal applications.

3.3.1. Earthen cover

ENRESA reported on the earthen cover design of the Spanish waste disposal facility El Cabril for the engineered service life of 300 years (Appendix 11). The performance requirements of the earthen cap is to resist erosion and continue to provide the following functional requirements:

- resist water infiltration into the waste disposal facility along with the concrete vault roof;
- resist intrusion of plants and animals into the waste disposal facility;
- resist inadvertent intrusion by humans;
- provide protection of the vault's concrete roof from the elements.

ENRESA concluded that the best research programme to validate the performance of the earthen cover for their disposal facility would be the surveillance of a mill tailings cap constructed in the Andujar Uranium Factory Closure Project, 80 km east of Cordoba. The mill tailings cap contains 8 layers of materials such as vegetation for soil protection, crushed rock to prevent erosion, top soil to facilitate vegetation growth, sand layer to facilitate drainage, cobble layer as bio-intrusion barrier, and clay as a barrier to radon gas and as a low permeable layer to restrict water infiltration.

3.3.2. Concrete barrier

Atomic Energy of Canada Limited (AECL) (Appendix 3) reported on their research programme on concrete durability. The objective of the research was to design a durable concrete for the required service life of 500 years for the IRUS facility for the disposal of the low level waste and to estimate the long term performance of a wide variety of concrete types and qualities under the disposal conditions. Four different water to cement ratios were used for the manufacture of concrete specimens to study the influence of this important parameter on the performance of concrete. Five different concrete systems with different combinations of the following cemetitious materials were selected for study:

- Ordinary Portland Cement;
- sulphate resisting cement;
- blast furnace slag;
- fly ash;
- silica fume.

The disposal environment was simulated by 25 baths containing various chemical ions and ionic combinations. Laboratory testing to determine diffusion rates of chemical species was initiated in 1988 and was still ongoing at the end of the CRP. Specimens were taken out of the baths at selected intervals and the diffusion fronts were located using scanning electron microscopes. Diffusion rates of chemical ions were evaluated for different qualities of concrete based on the depth of penetration of ions into concrete specimens with time.

AECL observed that the cement formulations containing supplementary materials such as blast furnace slag, silica fume and fly ash exhibit superior performance against chemical ingress compared to concrete made of Ordinary Portland Cement. Concrete formulations with lower water to cement ratios exhibited a decreased diffusion rate. Blended cements have also yielded superior performance with regards to permeability. The resistivity and conductivity experiments provided similar results, thus increasing the confidence in the test results. Based on the research methodology with reinforcement corrosion as the failure criteria and 75 mm of thickness of concrete cover to the reinforcement bars, AECL has concluded that the concrete formulation selected for the IRUS disposal facility (type 50 cement with slag and silica fume) would meet the required 500-year service life under the IRUS disposal conditions.

Research programme reported by ENRESA (Appendix 11) on concrete durability had the following objectives:

— design of a concrete composition to yield a durable concrete for the disposal vault;
— assessment of concrete longevity;
— demonstration of concrete performance under disposal conditions.

For the concrete formulations ENRESA used a minimum of 400 kg of cement content per cubic metre with a 0.42 water to cement ratio. To reduce the risk of reinforcement corrosion, the design of the structure was based on selected crack width criteria of 0.1 mm for concrete vaults and 0.05 mm for waste containers. The concrete surface was also provided with a 4 cm coating as an added protection. To assess the longevity, concrete samples from the disposal facility were tested for corrosion and carbonation. For the demonstration of durability under disposal conditions, instrumentation, non-destructive examination, and detailed modelling were used.

Bhabha Atomic Research Centre in India (BARC) (Appendix 9) reported that blast furnace slag concretes indicated higher chemical durability compared to concretes composed of Ordinary Portland Cement. They observed that slag-based concretes exhibited lower permeability and better leach characteristics and higher fluidity, aiding in the easier placement of concrete (workability).

Participants from Brazil (Appendix 2) reported that diffusion rate of chloride ions into concretes decrease with lower water to cement ratios.

The Nuclear Research Institute of the Czech Republic reported on durability of Dukovany disposal vaults (Appendix 7). It was reported that durability of reinforced concrete vaults is influenced by many factors, however, the dominant mechanism that may lead to the failure of structures would be the corrosion of reinforcement in the concrete roof panels. Based on the assumption that failure of the roof panels would occur when the area of the reinforcement bars are reduced by 50% due to corrosion, the longevity of the concrete panels was estimated to be 630 years including carbonation effects. Similarly, the service life of the vault walls was estimated to be 400 years.

ANDRA, participating as an observer at one of the RCMs, reported on the French research programme aimed at ensuring durability of concrete for the Centre de l'Aube LLW disposal facility with a service life requirement of 300 years. The research approach involved identifying the disposal environment, defining the performance criteria of concrete, formulation of a durable concrete, and quality assessment. A concrete structure designed for long term performance would require mechanical and chemical resistance. The research methodology involved an understanding of the degradation process, developement of theoretical degradation models, evaluation of degradation rate parameters and extrapolation of the results to 300 years. They have undertaken laboratory testing of cement pastes, fibre reinforced concretes slabs, concrete overpacks, and foundation concrete. The leaching studies have shown a degraded thickness of 1.4 mm in distilled water over 3 months or 4 cm extrapolated to 300 years. Freeze/thaw testing of rebar containers made of Ordinary Portland

Cement has indicated a 10 mm degraded thickness after 250 cycles. Similar tests on reinforced foundation concrete made of blended cement yielded a degradation thickness of 5 mm, indicating a required cover thickness of 4 cm for reinforcement protection. Major recommendations for concrete durability were:

- careful selection of cement (reduce portlandite as much as possible);
- maximum compactness;
- minimum water to cement ratio;
- special attention to curing;
- quality control and performance testing.

3.3.3. Other materials such as clay, vermiculite, bentonite

British Nuclear Fuels (BNFL) has reported on the in-situ performance of cut-off walls made of bentonite to limit the flow of water into and out of the Drigg disposal trenches (Appendix 12 and Section 3.2.3).

Natural clays achieve an equilibrium with the local groundwater that ensures that their permeability remains relatively constant. When used in the disturbed zone of a disposal facility, engineered and imported clays have not achieved this equilibrium and their permeability may be modified by the evolving chemistry of the disturbed zone. Examples of mechanisms that may change the permeability include dimensional changes in the clay lattice induced by ion replacement. At the Drigg site, both local clays from the site and engineered clays (bentonite – crushed limestone) have been used to provide barriers to groundwater infiltration. The extensive use of grout suggests that the clay may be affected by an alkaline aqueous phase. Tests were carried out with simulated grout porewater. The indigenous clay showed no degradation in performance. The permeability is variable for the clay samples used (6.4×10^{-8}–4.1×10^{-10} m/s), but remains unchanged when treated with an alkaline solution. The engineered clay shows an increase in permeability from 1×10^{-11} m/s to 6×10^{-11} m/s, but its performance remains superior to the natural clay. It was concluded, therefore, that these clay barriers will remain intact despite the perturbed chemistry of the disturbed zone.

3.3.4. Natural analogue studies on long-term performance of materials

The China Institute for Radiation Protection (Appendix 5) reported on the natural analogue studies on backfill materials from ancient Chinese constructions. Some ancient tombs (2500–5000 years old) were examined and were found to be in good condition and the contents well-preserved. They have observed a number of similarities between the ancient tombs in China and the near surface low level disposal facilities. Both require long-term isolation, both are man made, and are located near surface. These similarities provide the possibility that the understanding of techniques used in the construction of the ancient tombs would of value to the construction of disposal facilities. Also, since the ancient tombs are thousands of years old the timescale meets the service life requirements of the LILW disposal safety concept. Hence, these studies provide indirect but strong evidence that well designed engineered barriers can last for centuries in near surface disposal environment.

4. CONCLUSIONS

Disposal of LILW requires the use of barriers to control the isolation or release of radionuclides. In the absence of the natural barriers present in deep disposal, engineered barriers are an integral part of the strategy for near surface disposal facilities. The barriers must be considered during the design phase of new facilities but barriers are also used to improve the performance of operating sites or to remediate old or closed sites.

Selection of barriers is dictated by a variety of factors, such as local regulations and authorisation criteria, the nature of the waste, the local geology and hydrogeology, and the results from performance assessments of the disposal facility. To assist in this selection, data are required on the key properties of the barriers, for example, strength, permeability, and sorption capacity. In addition to the "as installed" properties of the barriers, details of their long term performance is also required.

This CRP focused on some of these issues. It should be emphasized that the output from the CRP is not comprehensive in terms of the barrier types, nor time-scales considered. The research programmes were directed at barriers that were specific to the needs of individual Member States that participated in the CRP. Based on the results and the discussions from the three RCMs, it is possible to draw the following conclusions:

- indigenous clays can provide an effective cap and backfill material;
- concrete structures will provide considerable structural integrity through the institutional phase of the disposal facility;
- horizontal groundwater flow can be restricted by use of cut-off walls;

It also became evident during the CRP that complex barrier systems such as caps must be considered as integrated systems. In addition, the use of mathematical models and associated computer codes to estimate the interactions between barriers and the surrounding geosphere should be encouraged to ensure the compatibility of the engineered barrier with local geology and hydrology.

Specific suggestions arising from this CRP included

- In view of the importance of barriers in near surface disposal, a comprehensive report on barrier properties should be produced;
- Special attention should be given to multi-layered cap systems;
- The development of mathematical models and associated computer codes to describe the interaction of barrier systems with geosphere should be encouraged.

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APPENDICES 1–13

EXTENDED SUMMARIES OF CONTRIBUTIONS FROM PARTICIPATING INSTITUTIONS

XA0200279

Appendix 1: BELARUS

Development of barrier composite coating technology for low level radioactive waste disposal

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This work is devoted to research relating to the development of new thin film barrier coatings, the associated preparation technique, and their use for solving various scientific and engineering problems associated with the improvement of the performance of low level waste disposal facilities. The work presents the main results of zirconium dioxide thin film coatings preparation process by organometallic composite solutions on various substrates and bases with subsequent thermal treatment at 400–1000°C.

Experimental work on the formation of isolating corrosion-resistant ZrO_2 (Y_2O_3) coatings over different substrates and bases, including microporous ones, indicate potential applications of the technology to coating barrier materials such as quartz, sand, concrete, alumina.

Two variants of pulse thermal treatment are proposed. The first variant was designed for samples with an irregular surface and having a large surface area. It is suggested that such surfaces are treated using step-by-step heating of local sections.

Experimental

1. Preparation technique

— $ZrCl_4$ alcoholised by ethanol with Zr $(OC_2H_5)_2Cl_2$ formation,
— Zr $(OC_2H_5)_2Cl_2$ dissolving at alcohol,
— dipping into the solution,
— drying in the air,
— thermal treatment in the furnaces for 1–10 seconds or 10–180 minutes at 200–500°C.

2. Deposition technique

Three glass ampoules were taken and 0.5 ml of $^{137}CsNO_3$ aqueous solution (1.6×10^6 Bq) were injected in each ampoule. After drying the ampoules under a lamp, 0.5 ml each of the protective solution were injected into two ampoules, which were dried under the lamp and annealed in a muffle furnace for 3 hours at 50°C. After cooling to room temperature, 0.5 ml of protective solution was injected into one of these ampoules again. The three ampoules were dried and annealed for 3 hours at 200°C. All the three ampoules were placed in beakers with 50 ml of tap water.

3. Measurements

Measurements of ^{137}Cs activity with a multichannel spectrometer gave the values shown in Table A.1, which are very small compared to the activity injected into the specimens.

TABLE A.1. ACTIVITY DYNAMICS OF ^{137}Cs (Bq) IN THE TAP WATER

Object under test	Conditions and time (hour) kept in the tap water					
	17.0		91.25		134.0	
	without ZrO_2	with ZrO_2	Without ZrO_2	with ZrO_2	without ZrO_2	with ZrO_2
Quartz	17600	1030	25200	10700	33600	21100
Sand	2140	338	6900	357	7530	355
Concrete	1120	210	1680	166	1770	234
Aluminium	38200	4220	35200	3510	37900	1980

Conclusions

The following can be concluded from the experimental test data:

- Relationships between the structure, chemical composition, physical and mechanical characteristics of oxide layers and organometallic compositions were established.
- Protective buffer coatings on microporous materials and formation processes were studied.
- The test results showed that zirconium dioxide protective films notably decrease radionuclides (^{137}Cs) penetration out of the samples into the environment. Moreover, the samples with two-layer zirconium dioxide coatings displayed more retention in comparison with one-layer samples.

XA0200280

Appendix 2: BRAZIL

Evaluation of engineered barrier materials for surface disposal facilities

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In practice, those nuclear installations that generate most wastes in Brazil often have interim storage facilities at the same site. They can also accommodate wastes from small users that have no suitable place to manage their wastes. The forecast at the time of the CRP for Brazilian waste generation due to all nuclear and radioactive activities by the year 2010 is to be about 5000 m^3, not including the waste of 3500 m^3 from Goiania accident. With the impact of the accident on the public opinion, the quest for a safe disposal facility became more urgent, especially the siting and licensing a disposal facility which could receive the Goiania waste.

Although the construction of a national repository was a matter of future decision at the time of the CRP, research programmes were being developed and carried out by the research institutes of the Brazilian National Commission of Nuclear Energy. The R&D programmes were primarily intended to establish required technical capability in dealing with the subject especially in terms of issues related to the evaluation of disposal facility performance and lifetime. Some activities of the programmes were within the scope of the CRP, namely:

- study of diffusion through cementitious materials;
- degradation of concrete due to chemical corrosion and microbiological attack;
- evaluation of additives for the improvement of structural concrete and cemented wasteform quality.

In the work relating to the improvement of wasteforms and concrete, silica-fume was being evaluated as an admixture in the cementation process. Ion-exchange resins, a typical power reactor waste, and simulated liquid waste from fission production were used as reference wastes. The performance of the produced wasteforms was evaluated by measuring four properties of interest: setting time; heat developed during hydration process, compressive strength, and leachability. Results showed that the addition of silica-fume increased the compressive strength for both type of wasteforms, the leachability of ^{137}Cs decreased even when the waste loading was increased. In the case of ion-exchange resins this increase could mean up to 50%. Analysing values of hydration heat and setting time, one can conclude that the addition of silica-fume does not alter those properties or at least has no negative influence on the process parameters and on the final product. As an admixture for concrete formulation, other relevant properties that were measured included: resistance to chemical corrosion (sulphate ions), permeability to gas, and diffusion of caesium.

With regard to the disposal facility for the waste from the Goiania accident, an IAEA/CNEN coordinated project was started in 1994 to provide technical support for the safety assessment. Support was provided by: training personnel; undertaking mathematical modelling, comparing test-case studies from the various institutes undertaking the assessments, and generating actual data through laboratory and field measurements.

XA0200281

Appendix 3: CANADA

Durability aspects of high-performance concretes for a waste repository

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The IRUS facility for the disposal of low level radioactive waste at the Chalk River Nuclear Laboratories in Ontario, Canada relies on the durability of concrete for the required 500 years of service life. A research programme based on laboratory testing to design a durable concrete and assess its long-term behaviour was initiated in 1988. This appendix discusses the methodology to assess the long-term behaviour of concrete, and some initial observations. Longevity predictions for concrete formulations based on diffusion testing are also presented.

The service life of concrete is dependent on a slow rate of deterioration and is influenced by the quality of concrete and the service environment. Factors such as cement type, cement content and water-to-cement ratio can affect the diffusion rate of ionic species into concretes. In addition, service life will depend on the failure criteria adopted. After examining the major failure mechanisms for the repository concrete, corrosion of reinforcement was selected as the mechanism for the failure of the waste repository structure. Chloride ions in the presence of oxygen can initiate corrosion of reinforcement and failure of the reinforced concrete components. The failure criteria chosen for the concrete was the time taken for the aggressive ions to reach the reinforcing steel by diffusion through the concrete cover (75 mm thick).

Experimental

1. Binders:
   Portland cement Type 10 and 50, silica fume and blast furnace slag were used for the three concrete systems described in this paper. The nomenclature and composition of cement systems are listed in Table A.2.

2. Aggregates:
   An unblended sand consisting mainly of quartz and feldspar was used. A limestone coarse aggregate of somewhat variable composition was used. Tests conducted were similar to those for the fine aggregate and the results were satisfactory.

3. Concrete:
   Three concrete systems (S1, S2, S5) were each prepared at four different water-cement ratios: 0.35, 0.42, 0.5 and 0.60, denoted as mix 1, 2, 3 and 4 (M1, M2, M3, M4).

The cement contents for S1, S2 and S5 are as follows: S1 (M (1-4)): 485, 370, 335 and 280 kg/m^3. S2 (M (1-4)): 383, 338, 338 and 275 kg/m^3; S5 (M (1-4)): 437, 359, 325, and 259 kg/m^3. A target slump of 125–150 mm was maintained for all mixes.

Two concrete prisms, 75 × 75 × 280 mm, were cast for each mix and each exposure condition. Prior to immersion in the test solutions, the prisms were coated with wax on all sides but one, to allow a unidirectional ingress of chloride or sulphate ions. On the basis of an analysis of the repository service environment, the following major degradation parameters were selected for laboratory testing of concrete specimens:

- sulphate ions, chloride ions, several agents in combination,
- leaching of calcium hydroxide by water, carbon dioxide reactions.

Ionic profiles and depth-of-penetration measurements (determined by EDXA) in concrete showed that reasonably accurate results can be obtained and predictions of ionic ingress made. There was some scatter in the experimental results, because of the difficulty of locating the reaction front in concrete test specimens, due to the tortuous path of ionic ingress through dense concrete. However, there was enough consistency and redundancy in the system to obtain fairly accurate results. The procedure following the diffusion path around the fine and coarse aggregate particles, using the scanning electron microscope and electron microprobe for analysis, was successful. The rate of penetration of aggressive ions into the concrete was evaluated by determining the reaction zone front with time of exposure in the solution baths. Prediction of long term concrete behaviour involves the extrapolation of current data, based on the assumption that long-term processes (not currently identified) will not invalidate the extrapolation. Figure A.1 shows the rate of diffusion of chloride ions into test concretes and extended regression lines for longevity predictions.

Conclusions

The following can be concluded from the experimental data:

- Hydrated blended cements mortars had diffusivities up to 25 times lower than equivalent. Type 10 hydrated Portland cement mortars and that the lowest diffusivities were obtained.
- Median pore diameter and $Ca(OH)_2$ content were ranked in the same order for the three cement systems (S5<S2<S1), and were similar to the ranking for electrical conductivity and diffusivity.
- Lower water-to-cement ratios in concrete systems decreased the diffusion rate of ions, and sulphate ions seemed to inhibit the rate-of-penetration of chloride ions.
- On the basis of the physical test results and the diffusion test data, System 5 concretes, with a 75% blast furnace slag replacing Type 50 cement and 3% silica fume blend, ranked the lowest with respect to permeability, and provide maximum resistance to degradation. System 5 mix 2 was selected as the candidate high-performance concrete for the repository construction. The experimental results indicated that this concrete would meet the 500 years service life requirement.

TABLE A.2. NOMENCLATURE AND COMPOSITION OF CONCRETE SYSTEMS

Concretes	Cement type	Blast furnace slag	Fly ash	Silica fume	Curing time
System 1 (S1)	Type 10 100%	0	0	0	14 days
System 2 (S2)	Type 50 90%	0	0	10%	14 days
System 5 (S5)	Type 50 22%	75%	0	3%	28 days

Mix 1 (M1) = 0.35 w/c
Mix 2 (M2) = 0.42 w/c
Mix 3 (M3) = 0.50 w/c
Mix 4 (M4) = 0.60 w/c

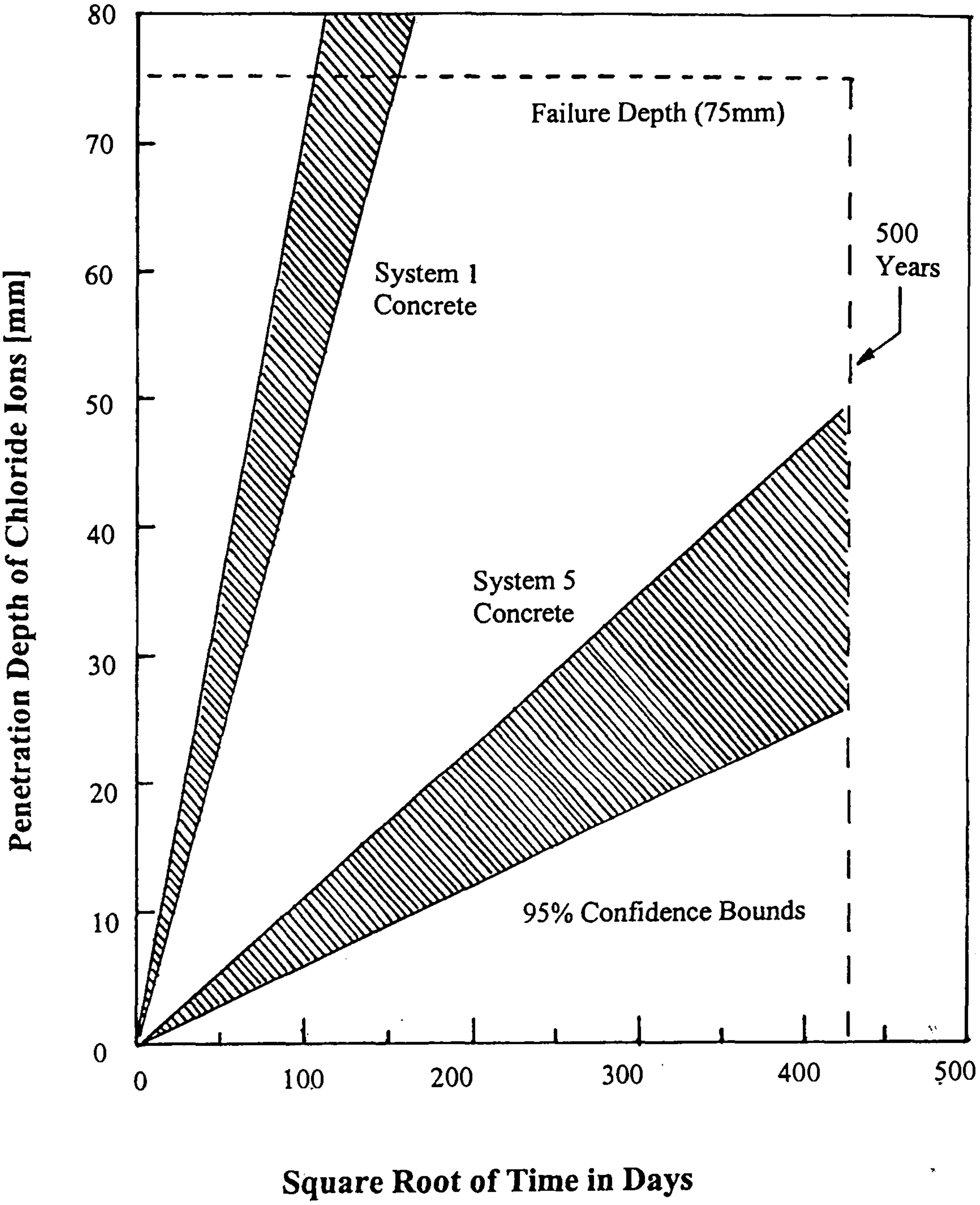

FIG. A.1. Extended regression lines; exposure of concrete specimens in 5 g/l chloride bath.

XA0200282

Appendix 4: CHINA (a)

Performance of backfill materials in near surface disposal facilities for low and intermediate level radwaste

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Backfill material is an important component of a multi-barriered disposal facility for low and intermediate level radioactive waste. This appendix describes the work concerning "performance study on engineering materials of shallow land disposal of low and intermediate level radwaste".

At the time of the CRP, China had planned to establish five regional disposal sites for low-and-intermediate level radioactive waste. According to the potential distribution of these sites, forty-three sampling points were selected through information survey and table discussion. After field survey and screening, eight of them were selected for further studies in laboratory. Basic physical and chemical properties of each sample were measured in laboratory. The results indicate that no one of the samples can individually function as the backfill material in a multi-barriered near surface facility. Then nine additives for adsorption modification were tested using a static method.

Further adsorption tests were conducted: three additives screened out in previous experiment were evaluated using the static method. Results obtained show that the Kd values of mixtures of 90% NW-3 and 10% BC for Co-60, Cs-134 and Sr-85, compared with those of 100% NW-3, are 4.8, 4.6 and 4.7 times higher, respectively. Effects of contact time, pH of tracer solutions and radionuclide concentrations of tracer solutions on Kd values of three samples, NW-3, BC and 90% NW-3 with 10% BC, were also be evaluated using the static method. Column tests were performed to evaluate migration of Co-60, Cs-134 and Sr-85 in NW-3 columns with different densities. The column tests were carried out for 210 days. However, no breakthrough was obtained.

Long term performance of backfill materials was assessed through natural analogue (see also Appendix 5). We compared Chinese ancient tombs with near-surface low and intermediate level radioactive waste (LILW) disposal facilities. Both were designed based upon multi-barrier principle. Then three backfill materials were collected from two Chinese ancient tombs in south China and an ancient architecture in northwest China and were studied in laboratories from the perspective of radioactive waste disposal in near-surface facilities. The results show that the two materials from the ancient tombs have low permeability and strong adsorption to radionuclide ^{60}Co and ^{134}Cs. The distribution coefficients of the two ancient materials for the two radionuclides were all in the order of 10^1 m^3/kg. The conclusion is that current LILW disposal option in the near surface would be effective for long term period of time, since clay materials are very effective in preventing water intrusion and retarding radionuclide release.

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Appendix 5: CHINA (b)

Natural analogue study on backfill materials from ancient Chinese constructions for LILW disposal

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The objective of this work was to contribute to the demonstration of the long term safety of low-and-intermediate level radioactive waste (LILW) disposal using information from a natural analogue study on ancient Chinese constructions.

The work firstly compared LILW near surface disposal facilities with Chinese ancient tombs in respects of siting, engineering structures, design and construction procedures and indicates that they are both based upon multi-barrier principle. After extensive literature and field survey, three materials were collected from two Chinese ancient tombs and one ancient architectures for further laboratory study.

The three materials were studied in laboratories from the point of view of radioactive waste disposal in near surface facilities to obtain information concerning their basic physical and chemical properties, engineering properties and radionuclide adsorption abilities. The results show that the two materials from the ancient tombs have low permeability and strong adsorption for ^{60}Co and ^{134}Cs. The saturated permeabilities of the two ancient materials are in the order of 10^{-10} m/s and the distribution coefficients for the two radionuclides are all in the order of 10^{1} m^{3}/kg.

The conclusion was that the then current LILW disposal option in near-surface would be effective for a long term period of time, and clay materials, as backfill materials for LILW near-surface disposal facilities would very effective in preventing water intrusion and retarding radionuclide release even over a long term of period. Overall the LILW disposal option was considered to be safe in long term.

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Appendix 6: CHINA (c)

Selection of backfill materials for the near surface disposal of low level radioactive waste

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As one of the engineered barriers for the near surface disposal of low level radioactive waste, the backfill materials may fulfill a number of roles, including minimization of groundwater access to the waste package, alteration of groundwater chemistry, and retardation of radionuclides transport. Over the past years, selections of backfill materials for radioactive waste disposal have been derived from a much data on adsorption behaviour of radionuclides on geological materials and the most widely advocated backfill material being bentonite. However, these selected backfill materials rely heavily on the cation exchange behaviour of the materials and do not contain other materials that have sufficient retardation capacity for some radionuclides such as ^{99}Tc and ^{129}I.

The behaviour of ^{99}Tc and ^{129}I in the geosphere is very important in low level radioactive waste management and disposal studies because of their long half-life and very low adsorption on most of the geological materials. Therefore, the aim of this work was to develop a multiple backfill material based on Na-bentonite for immobilising ^{99}Tc ,^{129}I and other mobile radionuclides, for a long period of time.

In order to identify geological materials that are capable of adsorbing caesium, strontium, technetium and iodine as the additional component of a multiple backfill material, the adsorption behaviour of Cs, Sr, Tc and I on Na-bentonite, Cs and Sr on gypsum, Tc and I on pyrite, magnetite, siderite, cinnabar, galena, stibnite, antimony ocher and tiemannite was studied. The experiments were performed in an aqueous phase of double distilled water by a batch technique at ambient temperature. The influences of pH, cations (K^+, Ca^{2+}, Fe^{2+}), groundwater, concentration of the nuclides on the adsorption of Cs, Sr, Tc and I were also studied, and the possible adsorption mechanisms were considered. According to the experimental results, several kinds of geological materials were recommended as the additional component of a multiple backfill material for enhancing its retardation capacity to Tc and I.

Experimental

The adsorption of Cs, Sr, Tc and I on Na-bentonite and the selected minerals was measured by a batch technique, with the experimental conditions as follows:

Solid phase	Na-bentonite and the selected minerals
Particle size	180 mesh
Aqueous phase	Double distilled water with variation of pH
Solid/Liquid ratio	1/10 (g/ml)
Radionuclide concentration	[^{137}Cs] = 2.6×10^{-7} M, [^{90}Sr] = 8.9×10^{-8} M [^{99}Tc] = 1.5×10^{-4} M, [^{125}I] = 3.3×10^{-9} M
Temperature	25 °C
Contact time	3 days
Solid/Liquid separation	Centrifugation, 0.5 h at 16 000 rpm
Experimental vessel	25 ml Polypropylene centrifugal tube with cap

A PE System 2000 NIR FT-Raman spectrometer was used to characterise the interactions of Tc and I with the selected minerals in this paper. The sample is 2.0 cm high, 2.0 cm wide and 0.1 cm deep.

Conclusions

The following results were obtained from the experiments:

- The adsorption behaviour of Cs, Sr, Tc and I on Na-bentonite and the selected materials seemed to be qualitatively related to the chemical properties of the radionuclides and the minerals.
- The adsorption distribution coefficient (Kd) value for Cs, Sr, Tc and I on Na-bentonite was 2.4×10^0 m^3/kg, 4.9×10^{-1} m^3/kg, 6×10^{-4} m^3/kg, and 3.3×10^{-3} m^3/kg, respectively. The influences of pH, cations, groundwater and concentration of the nuclides on the adsorption were different for the different nuclides. The adsorption mechanism of Cs and Sr on Na-bentonite was cation exchange reaction.
- The adsorption of Cs and Sr on gypsum was very poor, and this result was not in agreement with that reported before. Therefore, gypsum could not be included in the multiple backfill material for enhancing its retardation to Sr.
- The Kd value for Tc on stibnite, antimony ocher, tiemannite and magnetite is 2.6×10^1 m^3/kg, 2.3×10^1 m^3/kg, 1.5×10^1 m^3/kg, and 3.0×10^0 m^3/kg, respectively. The Kd value for I on tiemannite and galena is 6.2×10^0 m^3/kg and 3.5×10^{-1} m^3/kg, respectively. Therefore, stibnite, antimony ocher, tiemannite, magnetite and galena could be recommended as the additional component of the multiple backfill material for enhancing its retardation to Tc and I.
- FT-Raman analyses (Figure A.2) show that the adsorption mechanism of Tc on stibnite, antimony ocher, tiemannite and magnetite is redox reaction and the adsorption mechanism of I on tiemannite and galena is surface chemical adsorption.

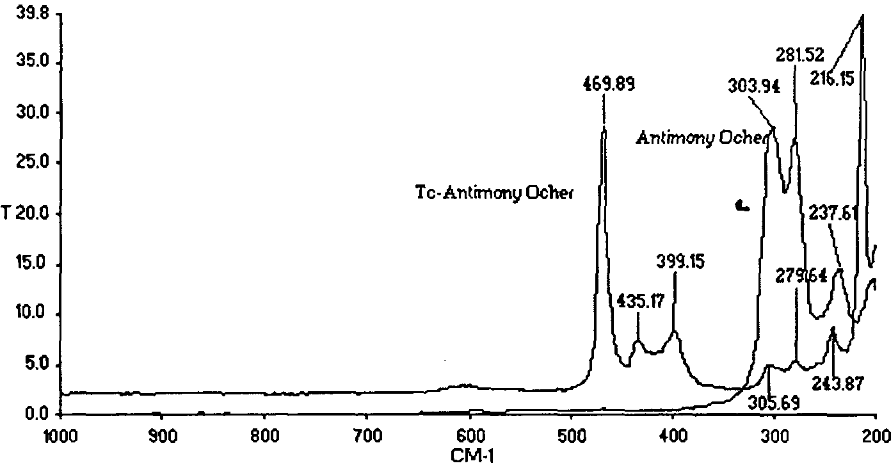

FIG. A.2. FT-Raman spectrum of antimony orcher before and after interaction with Tc(VII).

XA0200285

Appendix 7: CZECH REPUBLIC

Durability of Dukovany shallow land repository engineered barriers

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The main aim of this project was to explore the durability of engineering barriers used at Dukovany shallow land repository as a support of safety assessments.

This appendix summarises the principal results focused on durability of asphaltopropyleneconcrete (APC) hydroisolation and steel reinforced concrete construction.

Durability of asphaltopropyleneconcrete (APC) hydroisolation

One of the main disadvantages of hydroisolations based on asphalt is its organic components that are susceptible to microbial degradation. The study of durability of this barrier was therefore concentrated mainly upon this aspect of degradation.

Samples of microbial cultures of bacteria and fungi were repeatedly isolated from inside and the close surroundings of the Dukovany repository vaults (9.9.1992, 11.5.1993 and 5.12.1994) and both qualitative and quantitative determination of micro-organisms was carried out from water extracts of taken samples.

It was found that grampositive bacteria prevail in this region. On the surface of APC dominated the strain of Pseudomanas pseudoalcaligenes bacteria, various Micrococcus sp. strains and various Bacillus (G+).

The hydraulic conductivities of samples after radiation and microbial exposition were measured. A decrease of permeability after a longer term microbial activity was found. Isolated bacteria were incubated and inoculated to the cells with the microscopic glasses covered with thin layers of bitumen. After four months of cultivation it was possible even only by eye to observe the striking changes on bitumen samples.

The effect of temperature on the growth of some bacteria was studied at 5, 15, 30 and 40°C. It was detected that dominant biodeteriogen Pseudomonas Pseudoalcaligenes grows very well at 40°C , but can grow slowly even at temperatures about 5°C.

Metabolic activity of bacteria in the presence of bitumen and propylene oil, that forms the part of APC mainly from the reason of better workability of APC, was measured through determination of oxygen uptake by means of manometric method. The higher uptake of oxygen suggesting higher metabolic activity was achieved with bitumen samples.

On the surface of APC and concrete next to APC dominated the fungi of the family of Fusaria that were also present in the grass of the nearby slope. On the surface of concrete walls a higher extent of the fungi coming from soil or the surface of grass, namely Aspergillus sp., Alternaria or Clasterosporium was located.

The growth of isolated fungi on microscopic plates covered with bitumen was compared with the growth of the same strains of fungi on agar plates depending on relative humidity.

It was found that dominated Fusaria might grow at relative humidity of about 80%. (The relative humidity in some of concrete vaults may even exceed this value.)

The family of these fungi may also easily grow at temperatures about 5°C .

It is clear that neither temperature nor a lower humidity does not form a serious obstacle for growing of various kinds of fungi in concrete vaults.

A dramatic decrease of weight of bitumen samples grown in sterile soils inoculated by fungi and bacteria isolated at Dukovany Repository was observed. The change of weight was higher than the change of bitumen samples put in sterilized soil. This observation suggests the site specific and rather complex character of bitumen microbial degradation.

No definite conclusions from the results of the experiments, however, can be drawn for safety assessments. The problem is to quantify the microbial effects for real environment of Dukovany concrete vaults. The way forward is through long term field experiments and regular monitoring of possible microbial degradation throughout pre-closure period.

Durability of steel reinforced concrete construction

The durability of concrete vaults was estimated from the determination and comparison of concrete structure proportion in 1991 and 1995, i.e. 1 and 9 years after finishing of building.

The governing process that can lead to the failure of steel reinforced concrete walls is the corrosion of steel reinforcements mainly those that are in ceiling concrete panels.

The estimations made in this study were based on the assumption that the corrosion can only start after the thickness of the layer of the concrete above reinforcements amounts to the thickness of the carbonated layer, the possible effect of chloride ions has not been taken into account.

The layer above steel reinforcement was measured by means of electromagnetic indicator and the carbonated layer was estimated through measurements of change of pH by means of phenolphtalein indicator of drilled out samples.

From a comparison of results determined in 1991 and in this year it appears that carbonation process is slightly faster in walls than on the ceiling panels and is getting slower. For more realistic and more reliable results it will be necessary, however, to repeat measurements in longer intervals in future.

The rate of corrosion of steel reinforcement was estimated on the basis of empirical data collected for corrosion of uncovered concrete reinforcements in areas with various extent of pollution with sulfuric acid and air salinity.

On the basis of the results, obtained in the study, it was estimated that the total lifetime of the ceiling panels (including the carbonation interval and a decrease of steel reinforcements area to 50% at which the ceilings panels could with high probability sink down) is about 635 years.

XA0200286

Appendix 8: HUNGARY

Evaluation of silty clay as backfill and cover material for shallow land disposal facility in Hungary

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Hungary

As the last item of an intermediate term research programme connected to the Feke-Ofalu facility breakthrough measurements were carried out on local silty clay samples using HTO, I-125 and Co, Sr, Cs isotopes. The original aim of these measurements was testing of transport models in laboratory experiments and the study of the effects of inhomogenities in the material.

HTO and I breakthrough measurements were completed. Experimental method and apparatus were developed by Radiochemical Department of Veszprem University for elution type experiments, as well as for non-destructive scanning of columns for sorbing cations.

The main results of HTO and I measurements can be summarised as follows:

- The flow-regime in the columns was stable, no separation of fine fractions occurred, although the flow rate was determined by the frequency and amount of feeding;
- Neither HTO, nor I delay was observed on the breakthrough curves, which in fact showed no signs of sorption/inter-grain diffusion of these isotopes;
- 70–80% recovery in some cases represented a problem that remained to be solved.

As a preparatory work for the cation elution experiment, Kd measurements were performed for each soil sample, using carrier-free solution, 1M NaNO3 solution and 1×10^{-2} M solutions of the respective isotopes. The results showed reasonable decrease of sorption only for Sr isotopes.

The following further work was planned:

- Transport calculations would be performed for each sample and isotope, using the above mentioned Kd values;
- Elution and scanning would subsequently be performed for each column, aiming to detect movement of sorbing cations;
- At the end of experiments, distribution of sorbing isotopes would be measured by destructive methods.

Hydrogeological and transport modelling in the final cover designed for Feled-Ofalu facility would be carried out to test the modified models.

XA0200287

Assessment of multiple barrier system components for waste isolation in near surface disposal facilities

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Naturally occurring backfill materials studied and characterised included vermiculite, bentonite, attapulgite and clayey soils from different sources within India. Apart from physico-chemical characterization, studies included radionuclide sorption behaviour of the backfill materials and their desorption in groundwater.

While adequacy of Ordinary Portland Cement (OPC) for immobilisation of low level waste is well established, several new cement blends were specially formulated and tested to be compatible for intermediate level waste. These wastes are highly alkaline and had higher salt loading especially nitrates and nitrites. Screening of these formulations was carried out to select the most promising formulations which were further subjected to detailed investigations with respect to physico-chemical characteristics and long term leach behaviour.

At the time of the CRP, various methods were under study for the top sealing/closure of trenches after disposal of waste. These included natural soils, reinforced cement concrete slabs in combination with cement mortar and brick-bat-coba, crevice sealing materials like bitumen, polymer, cement grouts etc.

Experimental

(a) Backfill materials

Sorption studies for Cs-137 and Sr-90 on vermiculite and bentonite were carried out in the presence of competing ions (Na^{+1} and Fe^{+3}). Studies on evaluation of admixtures comprising of these backfill materials (vermiculite, bentonite) and sand in different proportions were carried out as well. Individual sorption for Cs-137 and Sr-90 using batch equilibration method was about 35 meq/100 g for vermiculite and about 53 meq/100 g for Sr-90. Table A.3 gives the admixture proportions of various backfill materials. The values for sorption capacities for these admixtures are given in Table A.4.

TABLE A.3 ADMIXTURE PROPORTIONS

Admixture components	Set 1	Set 2	Set 3
Vermiculite	0.4	0.3	0.3
Bentonite	0.3	0.4	0.3
Sand	0.3	0.3	0.4

TABLE A.4 SORPTION OF Cs-137 AND Sr-90 ON ADMIXTURES

Sorption Capacity (meq/100g)			
	Set 1	Set 2	Set 3
Cs-137	31.0	33.0	27.0
Sr-90	19.0	16.2	14.9

(b) Slag-based cements

Detailed experimental studies for slag-based cement formulations, as a waste form, indicate that hydration of these cements is much slower compared to Ordinary Portland Cement. Scanning electron microscopic examination revealed Calcium-Silicate-Hydrates forming inter-granular bridge between slag particles with incipient precipitation of C-S-H on their surfaces. On comparison with OPC, the percentage of unhydrated cement was found to be much lower in slag-based cements. The size of the unhydrated particles was also smaller compared to OPC.

(c) Engineered structures

Detailed monitoring provisions were made outside the trenches (near surface engineered facility for storage/disposal of solid and solidified waste) to attain the direction of groundwater movement. The effectiveness of the system leak tightness and integrity was evaluated by having inspection pipes connected up to the bottom sump of the trenches. Outside the containment walls, a water collection network of perforated pipes at raft level was provided leading to a sump for monitoring radioactive contamination. Lastly, a grid work of boreholes provided data on migration of radionuclides, if any, by periodic sampling and analysis of groundwater.

(d) Sealing/closure of engineered structures

Sealing was provided for the earth trenches by covering them with a sufficiently thick layer of excavated soil, which was allowed to consolidate by sprinkling water. The void space in the trench was also filled with soil and vermiculite mix. The top of the reinforced concrete trenches/vaults was closed with precast concrete covers. After placement of waste packages, in some of the cases, top was filled with backfill materials like soil, vermiculite, crushed stones, cement grout etc. The crevices between concrete slabs were sealed with bitumen, polymer and cement grout. A study on the various waterproofing/sealing procedures was undertaken. A system under evaluation consisted of a RCC slab followed by polythene sheet, rich cement mortar, brick-bat-coba and plastering with water repellent cement.

Conclusions

Admixture formulations of backfills indicate that with increasing content of high exchange capacity of the mineral present in the admixture, the sorption of Cs-137 increases. Presence of a higher content of vermiculite resulted in more effective removal of Sr-90.

Special slag-based cements appear to be a potential candidate matrix for conditioning of highly-alkaline and high-salt loaded intermediate level wastes. It also served as an excellent backfill material to be used in near surface disposal facilities. Its physico-chemical properties and mechanical strength were superior to Ordinary Portland Cement. The cumulative percentage activity released was 0.159 percent of the total activity over a period of 655 days for intermediate level wastes, having activity in the range of 1 Bq/m^3 and a salt loading of nearly 300 kg/m^3.

The Kd values of the soil used as a backfill around trenches/vaults being very high (1–3 m^3/kg for Cs-137), it is observed by ground water monitoring that migration of activity is practically nil over a long period in the direction of ground water. The borehole activity analysis for a typical study has confirmed that activity at a distance of 1 meter from the source term (3.7×10^{-6} to 3.7×10^{-7} Bq/m^3) has remained steady at 3.7×10^{-8} to 3.7×10^{-9} Bq/m^3 level over a monitoring period of 10 years.

XA0200288

Appendix 10: REPUBLIC OF KOREA

Use of the mixture of clay and crushed rock as a backfill material for low and intermediate level radioactive waste repository

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Background

At the time of the CRP, a repository for low and intermediate level radioactive wastes arising from nuclear power plant operation and radioisotope application in the Republic of Korea was to be constructed in the bedrock below ground surface. As the intermediate level waste cavern would contain the major part of radionuclide inventory in the cavern, the radionuclide release from the intermediate level waste cavern was therefore important from the viewpoint of disposal facility performance. The then current design concept suggested that the intermediate level waste would be emplaced into the compartment made of reinforced concrete, and the space between the concrete wall and cavern surface would be backfilled with a clay-based material.

As compacted clay-based materials have a low hydraulic conductivity and the hydraulic gradient in a disposal cavern was expected to be relatively low, molecular diffusion was considered to be the principal mechanism by which radionuclides would migrate through the backfill. The mixture of calcium bentonite and crushed rock was being suggested as a candidate backfill material.

This appendix summarises the KAERI research activities on the evaluation of hydraulic conductivity, radionuclide diffusion coefficient, and mechanical properties of the candidate clay-based backfill material for the intermediate level waste cavern.

Experimental

1. Materials

The clay being considered for the backfill material was a calcium bentonite from Younil, Kyungsangbukdo, the Republic of Korea, and it contains approximately 53.2% SiO_2, 22.1% Al_2O_3, 8.4% F_2O_3, 2.6% CaO, 2.0% MgO, 1.4% Na_2O, and some minor elements. Only clay portion that passed a No. 200 size sieve (with opening of width 0.074 mm) was used. The rock aggregate was crushed granite from Daeduk, Taejon. The maximum and minimum particle sizes of rock aggregates were set at No.4 sieve size (with the opening of width 4.70 mm), and No.200 sieve size, respectively.

2. Hydraulic conductivities

To measure the hydraulic conductivities of clay and crushed rock mixture, distilled water was used for the sample preparation and subsequent testing. The apparatus used to determine the hydraulic conductivity was designed to supply water into the sample at the hydraulic pressure of 1.3 kg/cm^2. The cylindrical chamber was 5 cm in diameter, and 5 cm in height, and the distilled water flows from the bottom to the top of the sample chamber at room temperature. The penetrated water volumes were measured by weighing.

3. Diffusion coefficients

The clay was saturated with a synthetic groundwater solution (SGW) with a pH of 7.0 and the following composition (in ppm): Na, 8.3; K, 3.5; Mg, 3.9; Ca, 13; Cl, 5.0; SO_4, 8.6; NO_3, 0.62; F, 0.19; HCO_3, not detectable. Both "in-diffusion" and "through-diffusion" methods were used to measure diffusion coefficients in compacted saturated clay. The clay was compacted to a dry density of 1.4 Mg/m^3, and the SGW was spiked with ^{90}Sr, ^{137}Cs, ^{60}Co, and ^{125}I.

4. Mechanical Properties

For the mechanical properties, Atturberg limits, the compaction property, the compressive strength, and the consolidation property were measured.

Conclusions

The hydraulic conductivities of clay/crushed rock mixtures decreased with increasing clay content. When the clay content increased to 50 wt%, the hydraulic conductivities of mixtures maintained the lower values, about 3×10^{-10} m/s, even at the dry density of 1500 kg/m^3. When the dry density increased to 2000 kg/m^3, the hydraulic conductivities decreased considerably, and were below 1×10^{-10} m/s, at the clay content of wt% so that the principal mechanism of radionuclide transport through the proposed backfill material would be molecular diffusion.

The diffusion coefficients of ^{137}Cs, ^{90}Sr, ^{60}Co, and ^{125}I in the clay with dry density of 1400 kg/m^3 were similar to those for Na-bentonite. The apparent diffusion coefficients in the clay with dry densities of 1000, 1200, 1400 and 1700 kg/m^3 decreased with increasing clay dry density, and the values were in the range of 3.80×10^{-11} to 7.12×10^{-11} m^2/s for ^{125}I, and of 1.21×10^{-12} to 1.43×10^{-12} m^2/s for ^{90}Sr. The values of apparent diffusion coefficients were not sensitive to the clay dry density, and varied by less than a factor of two.

Atturberg limits increased with increasing clay content and the clay/crushed rock mixture showed considerably good volume change potential. The unconfined compressive strengths of the clay/crushed rock mixture were in the range of 195 kN/m^2 to 269 kN/m^2 at the clay content of 20 wt% to 40 wt%. The consolidation coefficient at the consolidation pressure of 80 kN/m^2 to 320 kN/m^2 were 0.77 m^2/y to 1.65 m^2/y for clay content of 20 wt%, 0.88 m^2/y to 1.15 m^2/y for clay content of 30 wt%, and 0.18 m^2/y to 0.36 m^2/y for clay content of 40 wt%.

XA0200289

Appendix 11: SPAIN

Performance of engineered barriers materials in near surface disposal facilities in Spain

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Background

In October 1992 the Ministry of Industry and Energy issued the Operating License of El Cabril Near Surface Disposal Facility, in the province of Córdoba, some 100 km away from Córdoba city. Waste packages, mainly 0.22 m^3 steel drums, containing solidified waste in a cement based waste form or pellets coming from the super-compaction process, are placed inside concrete disposal containers. These containers are made of reinforced concrete and in their construction fabrication joints have been avoided. Once these containers are filled with 18 drums (0.22 m^3) or 30 to 60 compaction pellets, they are backfilled and sealed with a mortar grout, resulting into a solid block.

These blocks are then disposed of inside concrete vaults, called disposal cells, each one with a capacity for 320 containers. The full vaults are backfilled with gravel in the existing central gap left to absorb fabrication and handling tolerances. Then a plastic film is placed on the containers to prevent a true union between the last layer of disposal containers and the massed concrete layer cast to protect the workers during the construction of the closing slab. This 0.5 m thick closing slab is made of reinforced concrete and is protected by acrylic/fibreglass unperceived film. Galleries are made of a 300 kg/cm^2 characteristic strength concrete.

Concrete used in vaults and containers were upgraded to 350 kg/cm^2 characteristic strength. The concrete used in vaults construction and containers fabrication is the same, the only difference being the maximum aggregate size, limited in the case of containers to 16 mm.

For the performance of the final earthen cap, the best research programme was the surveillance of the mill tailings cap constructed in the Andujar Uranium Factory Closure Project, some 80 km east of Córdoba.

The concrete used in the construction of containers, vaults and galleries was defined after a research programme established to optimise their durability. The research programme can be divided into three major steps:

- optimization of durability,
- useful lifetime assessment,
- demonstration under disposal facility environmental conditions.

Optimization of durability

El Cabril presents favourable conditions regarding durability of concrete structures, due to the following reasons:

- It is far away from marine environment (absence of chlorides in air or solid deposits).
- Itis in a rural area without air pollution.

- Site underground water and soil, as well as dam water, used in concrete preparation do not present significant quantities of chloride, sulphate or other aggressive.
- Climate is mild with no special frost hazards.
- The disposal system is located above water table level.

The possible degradation due to an expansive alkali-aggregate reaction has been avoided by means of a careful selection of materials. On the one hand by the use of a low C_3A (Tricalcic aluminate) content cement. On the other hand siliceous aggregates of the area were selected, after a control by standard reactivity tests and by × ray diffraction to determine major mineralogy compounds. This required, for instance, well characterised aggregates of El Cabril area to be sent to a factory for the construction of the first 100 containers, for the proposed aggregates presented in the × ray test a dolomite content, after having passed successfully the conventional standard test.

Even though, as indicated, the site water and the concrete mixing water were free of aggressive, a sulphate resistant-sea water resistant cement was selected. Three cement factories were pre-selected, retaining the one with lower air permeability and lower carbonation coefficient.

In order to have a compact concrete to minimise progress of aggressive intrusion or carbonation progression the design characteristic strength was upgraded from 300 kg/cm^2 to 350 kg/cm^2. A minimum portion of 400 kg of cement per cubic meter was specified to have a reserve of alkalinity. The Spanish standard designation of the cement used is I 45 A SR MR (Portland, 450 bar min. characteristic strength, resistant to sulphates and sea water). Water/cement ratio was limited to get a low permeability to 0.42.

Influence of micro-cracking was also studied. For cracks of 0.1 to 0.2 mm no increase of bars corrosion was observed. Crack size was limited by design to 0.1 in vaults and to 0.05 mm in containers.

As important as the materials specifications is the control of their quality. During design and construction, and for containers fabrication, a quality assurance system was implemented, with an intensive control. During construction of the disposal cells some 28,000 probes were made. The main strength obtained was 450 kg/cm^2 with a standard deviation of 22 kg/cm^2 thus denoting a good quality and homogeneity.

With the cautions adopted, the major risk is the corrosion of reinforcement bars, after the loss of passivation of iron because of the decrease of pH produced with the carbonation of concrete. In addition of the above mentioned use of a very compact concrete, a coating thickness of 4 cm, the maximum admitted by Spanish construction rules (EH 92), was incorporated into the design.

Degradation by biological action can only be significant in media favourable for bacterial build up. This can be neglected in El Cabril conditions. Concrete structures aged more than 40 years, including a large dam, were studied. Good behaviour was observed in all cases.

A selection and characterisation of the backfilling grout was also developed in this phase. The mortar structural requirements were less important, but it presented additional functions to fill: interface with waste forms, low heat of hydration, no retraction, and an enough adherence to the structural concrete. A fly-ash puzolanic cement was selected (Spanish standard designation IV 35 A SR MR BC).

In this phase studies were carried out on alternative materials for the containers fabrication, such as metallic glass fibre. The alternative was not agreed because there were some doubts about its performance in very alkaline environments.

Lifetime assessment

In the first phase of studies, the significant processes of degradation determined were leaching (for concrete), and carbonation of the coating, mainly in the period in which the structures are uncovered (for reinforcement bars).

In the first case there was a progressive loss of mechanical capacity of the structure. When the degradation arrived at the reinforcement bars, these began to corrode, producing an acceleration of the degradation.

In the case of bars corrosion produced by carbonation or chloride intrusion, both chlorides and carbonation were almost innocuous for concrete, thus there was no significant degradation of the assembly till these processes reached the reinforcement bars (initiation period). As in our case the carbonation did not reach the reinforcement in the 300 year period considered in the model, it was not necessary to assess the propagation period in which there was a loss of the section of the bars.

For the assessment of the carbonated depth a simplified solution of proportionality to the square root of time was adopted. The carbonation thickness was measured in different probes, some of them two and a half years old, obtaining carbonation coefficients of 1–2 mm/$y^{0.5}$. Carbonation progress for 10, 20, and 300 years (foreseen duration of operational phase, before capping of different zones, and maximum extent of surveillance period), assuming the worst estimated value of the carbonation coefficient (2 mm/$y^{0.5}$) was calculated, resulting depths of 6.5, 9, and 35 mm. In the last case it was supposed that real depth should be smaller, because carbonation advance should be much slower or interrupted after site closure. As the calculated carbonation thickness was smaller than the actual covering thickness, it was thought that no important degradation would be found in that period.

Leaching needs a flow of water. For the expected hydraulic conductivities of 10^{-11}–10^{-12} m/s water front progress in 300 years would be lower than 100 mm, the leaching effects in that period should be of insignificant effects. Loss of mechanical strength was not considered to be important, due to the structural allowances.

Another way of water intrusion is capillary absorption. Capillary absorption coefficient measured up to 13.22 mm/$y^{0.5}$. Water penetration in 300 years by capillary absorption coefficient would be around 230 mm. This means that no important leaching effects were expected in this case.

R&D plan on durability under disposal conditions in El Cabril

During the CRP, a the third phase (assessment of the durability under actual disposal conditions) was started whose goal was to obtain a good estimate even for periods longer than 300 years, after the collection of data evolution of the parameters that control the ageing of the structures. This Plan was divided in four parts:

- collection of data from an instrumented disposal container stored in the same way they were disposed of inside the vaults;
- non-destructive examination of the constructed disposal vaults;
- destructive test on probes coming from the construction of vaults and from fabrication of containers;
- detailed modelling.

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Appendix 12: UNITED KINGDOM

Engineered barriers at the low level waste disposal site at Drigg in the United Kingdom

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The Drigg Low Level Waste Disposal Site has been in operation since 1959. The disposal concept initially used was tumble tipping into trenches cut into the host geology. This practice was continued until seven trenches, taking in excess of 800,000 m^3 of waste, had been filled. At this stage the trenches were covered with a low permeability cap to limit rainwater infusion.

In the 1980's a government committee reviewed the operations on site. Although the site was forced to operate within the requirements of the authorisation, a series of improvements were implemented in line with the recommendations of the committee's report. These included interception of leachate for discharge through a marine pipeline and the adoption of the vault concept. This concept involved compacting and grouting waste and emplacing the waste in the concrete vault. On filling the vault, a low permeability structure will be installed to limit infiltration of rainwater.

To support these and other improvement on the Drigg site an experimental programme was initiated. Some of the results of this programme are presented in this appendix.

The programme itself evolved over the period of the CRP and a model, the Engineered Barriers Model, was developed to explore the complex interactions of the various barriers and migration pathways for radionuclides. Much of the work moved from a semi-empirical study of individual barriers to validations of families of barriers used to control radionuclide release.

At the completion of the CRP, the Engineered Barrier Programme consisted of:

- assessing the performance of the site as it then existed by providing data on the barriers extant;
- defining cap options;
- providing support for future development of the site.

This appendix presents some of the results using the validation rigs plus some of the work initiated early in the programme.

The validation rigs were used to investigate rates of infiltration and lateral flow against Drigg Rainfall (ca. 0.75 m/year), the effect of sequential drainage and low permeability layers and the effect of cap settlement. Trials demonstrated that for a low permeability clay (4×10^{-8} m/s to 1×10^{-8} m/s) used in sequence with a sand-soil-kaolin drainage layer the cap at Drigg could shed the annual rainfall burden and could accommodate a settlement of 500 mm.

A barrier type that is used at Drigg to control horizontal groundwater flow is the cement-clay cut-off wall (bentonite and Ordinary Portland Cement with Ground Granulated Blast Furnace Slag). This barrier is keyed into a low permeability clay horizon. The bulk permeability of the wall material itself is 1 × 10^{-9} m/s but field trials using a cell set into the wall showed a much higher, though nonetheless acceptable permeability of *ca.* 1 × 10^{-8} m/s. This higher value was thought to be a feature of engineering difficulties in sealing the vertical wall with the underlying strata.

Engineered clay has been used to augment existing natural clay. This clay (bentonite and crushed limestone) may be susceptible to changes in groundwater chemistry and subsequently show a degraded permeability. Two sets of measurements were carried out. The first was to measure the effect of an alkaline solution on the clay. The natural clay showed no change in permeability however the engineered clay showed a marginal increase in permeability under forcing conditions (1 × 10^{-11} to 6 × 10^{-11} m/s). The engineered clay therefore still performed favourably compared to the natural clay. In the second study the evolving chemistry beneath the Drigg Vault was measured. No evidence was detected to suggest a significant increase in groundwater alkalinity which suggested the low permeability of the emplaced clay would be sustained.

Trials were completed on sorption of nuclides onto barrier materials although it was felt unlikely that the cut-off wall, engineered clay or structural concrete would significantly contribute to nuclide retention compared to the waste itself and the in-fill used in the waste form.

To establish the leach rate some long term leach tests were established. These test involved grouted, compacted waste and had the aim of establishing leach rates, probate chemistry and degradation of waste (corrosion and degradation of organics). The long term leach test samples were only beginning to 'settle', i.e. reflect the unperturbed samples, at the end of the CRP.

The CRP was also offered the following publications:

[1] D.P. Trivedi, R.G.G. Holmes and D Brown, Monitoring the In-Situ Performance of a Cement-Bentonite Cut-Off Wall at a Low Level Waste Site, *Cement and Concrete Research*, Vol 22, p339, Pergammon Press (1992).

[2] D.P. Trivedi and R.G.G. Holmes, Monitoring the Evolution of the Clay Environment Beneath a Concrete Vault at the Drigg Low Level Waste Disposal Site, *The Geochemistry of Clay-Pore Fluid Interactions,* The Mineral Society, London (1991).

[3] C J Stafford, S Richardson, D P Trivedi, Radionuclide Sorption Studies in Support of the Drigg Low Level Waste Disposal Site, *Migration '95*, St Malo, France (1995).

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Appendix 13: USA

Innovative technologies for the remediation of transuranic-contaminated landfills

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Given the complexity and heterogeneity of buried waste associated with the United States Department of Energy, identifying appropriate remediation schemes has required considerable effort. Preliminary evaluations indicated that significant technological advancements were required to safely and cost-effectively complete the remediation of Department of Energy buried waste sites, particularly within established timeframes.

The Transuranic-Contaminated Arid Landfill Stabilization Programme, formerly the Buried Waste Integrated Demonstration Programme, was organized by the Department of Energy, Office of Technology Development, to (a) manage the development of emerging technologies that could be successfully applied to remediation and (b) promote the use of these technologies to improve environmental restoration and waste management operations for transuranic-contaminated landfills in arid environments.

Implementing the Transuranic-Contaminated Arid Landfill Stabilization Programme involved three key strategies: 1) A systems engineering approach was used to include an overall perspective of the entire remediation process; 2) State-of-the-art science and technology were sought for improving the remediation system; 3) Integrated product teams which were comprised of end users, regulators, stakeholders, as well as industry partners were formed.

The Transuranic-Contaminated Arid Landfill Stabilization Programme focused its resources on various remediation technology systems. The In Situ Stabilization System and the Selective Retrieval System were two innovative systems that were directly applicable to the CRP.

The In-Situ Stabilization System involves the encapsulation of previously disposed waste. A jet grouting technique operating at 6000 psi was the emplacement technique used for these field experiments. This system showed that improved stability to the waste form was achievable if the waste cell was solidified. This approach reduced the hydraulic conductivity of the disposal cell to 2.8×10^{-14} m/s thereby reducing both surface and groundwater intrusion. Researchers also:

- investigated the spread of contaminants during the injection and removal operation,
- determined the effectiveness, and durability of grouting a soil/waste matrix,
- assessed the retrievability of a grouted soil/waste matrix,
- investigated grout emplacement techniques for high clay-content, low permeability soils, and
- investigated improved waste forms such as vitrified, cementitious and polymer grouts.

Results showed that in situ stabilization can prevent migration of contaminants from the buried waste. In situ stabilization can also provide an additional means of structural support for caps and surface barriers. Both interim and long term stabilization materials were investigated to provide remediation alternatives to satisfy a variety of site characteristics.

The Selective Retrieval System was an example of another type of technology system that can provide an indirect barrier. This system involves the use of remote selective retrieval equipment, including advanced waste handling, retrieval, and conveyance techniques. Using remote operations, operators can safely and efficiently remove buried waste contaminated with radioactive or other hazardous constituents. These techniques remove the workers from the hazardous and radioactive contaminants while permitting segregation of various waste types for conditioning, repackaging and improved management. Results of this research were still ongoing at the end of the CRP. Preliminary studies showed that production rates may be as low as 50% of the baseline manual techniques. However, safety and factors were significantly enhanced through the use of remote systems.

The Transuranic-Contaminated Arid Landfill Stabilization Programme supported environmental remediation efforts by seeking out the best talent to solve technology challenges identified in baseline remediation strategies. Experts from throughout the Department of Energy, universities, private sector, and international community were included in the programme to ensure implementation and commercialisation of innovative technologies.

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