

NDA Report no. DSSC/452/01

# Geological Disposal Engineered Barrier System Status Report

December 2016





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# Abstract

The Engineered Barrier System status report is part of a suite of eight research status reports. The purpose of the research status reports is to describe the science and technology underpinning geological disposal of UK higher activity wastes by providing a structured review and summary of relevant published scientific literature and discussing its relevance in the UK context. The reports have been written for an audience with a scientific or technical background and with some knowledge of the context of geological disposal. The current suite of research status reports (issue 2) updates and replaces the suite produced in 2010 (issue 1).

The objective of the Engineered Barrier System status report is to summarise the scientific evidence relating to evolution processes of the EBS during periods of storage and after disposal in the GDF. The key message emerging from the analysis presented in this status report is that it is important to understand which key processes affecting evolution of the engineered barrier system are likely to occur, how important these processes are for different disposal concepts and different geological environments, and the order and timescales over which they occur.

# DSSC/452/01

# Executive summary

The Engineered Barrier System status report is part of a suite of research status reports describing the science and technology underpinning geological disposal of UK higher activity wastes.

The report sets out RWM's understanding of the long-term evolution of the engineered barrier system (also sometimes known as the 'near field') of a Geological Disposal Facility (GDF). This requires knowledge of how different engineered barriers can help to prevent or limit the release of radionuclides and their migration to the host rock, through understanding and demonstrating the functions that different barriers provide as part of an integrated multi-barrier disposal concept. The engineered barriers may also provide functions in support of operational activities and worker health and safety. However, these aspects are not fully considered here.

The understanding of the evolution of the engineered barrier system described in this report draws on a number of sources, including work undertaken by ourselves (and previously that carried out by United Kingdom Nirex Ltd), in addition to information available from other organisations that are investigating the possibility of developing a geological disposal facility for the disposal of radioactive wastes (such as regulators, other waste management organisations and research institutions worldwide).

To demonstrate how the engineered barrier system (including the wasteform, waste containers, buffer materials, backfill, and seals) contributes to the safety strategy of the GDF over post-closure timeframes, it is necessary to consider all aspects of its evolution. Some of these barriers (for example, the waste package) are the subject of other reports. In such cases the discussion is limited to a summary of key processes occurring, together with an evaluation of how coupled interactions between different components of the engineered barrier system affect its overall evolution. Cross references to more detailed discussion in other status reports are provided where relevant.

The nature of the host rock and the groundwater infiltrating the engineered barrier system will have a significant influence on how it evolves, and the impact of these components of the disposal system on the engineered barrier system needs to be considered. Three host rocks are considered as the basis for discussion: a higher strength rock, a lower strength sedimentary rock and an evaporite rock. We consider the engineered barrier systems in two main disposal areas. This includes one disposal area for principally high-level waste (HLW) and spent fuel, and a second for intermediate level waste (ILW), including a small fraction of low-level waste (LLW) that cannot be disposed of at the Low Level Radioactive Waste Repository near Drigg in Cumbria. Covering three host rocks, this leads to a discussion of six illustrative concepts.

Typical processes dominating the evolution of the engineered barrier system applicable to the range of illustrative disposal concepts considered include those processes occurring during the construction and operational period, such as desaturation and oxidation of part of the host rock immediately surrounding the GDF.

Once wastes and the buffer and/or backfill materials have been emplaced and disposal areas have been sealed, a series of processes will begin during the early post-closure period that may then continue throughout the late post-closure period. These include:

- heat generation from the emplaced waste (and, to a lesser extent, from exothermic reactions such as those involved in the hydration of cementitious materials) and subsequent thermal effects
- irradiation of engineered barrier materials (particularly of waste package components)

- hydraulic saturation this evolution of the engineered barriers may include swelling of clay buffer and backfills, and hydration of cement buffers and backfills
- host rock creep, resulting in the compaction of clay-based and crushed salt buffers in poorly indurated clays and evaporites, respectively
- gradual degradation of engineered barriers, including buffers, backfills and seals, by various processes under lower temperature conditions and in less intense radiation fields, leading to long-term evolution of barrier properties and porewater chemistry
- consolidation and evolution of the disposal tunnels and shaft seals and plugs
- gas generation due to corrosion, microbial degradation of some organic materials and radiolysis of water and organic materials
- microbial effects, such as corrosion and other degradation effects
- waste container evolution, including corrosion in disposal concepts where water is present.

The extent and chronology of these processes will vary between different concepts. However, in all cases, they are likely to overlap to an extent, and some would be coupled to varying degrees. The specific processes occurring within the evolving engineered barrier system of the GDF will depend upon a number of factors, including:

- the nature of the waste inventory, in particular:
  - whether it will generate significant heat through radioactive decay
  - how the waste is conditioned to produce a passive wasteform, and how this wasteform is likely to behave over the long term
  - whether waste degradation (and container corrosion) is likely to cause significant gas generation
- the choice of engineered materials used, and how these will interact with each other, with the waste and with the surrounding host rock
- the nature of the geological environment and the hydrogeological conditions.

The engineered barrier system provides long-term containment of radionuclides, thereby limiting their release to the geosphere. Over time the barriers will evolve, as they interact with each other and with their surroundings. In combination with the continuing containment and isolation provided by the geosphere, the engineered barrier system will contribute to the long-term safety provided by the GDF as part of our multi-barrier approach to geological disposal.

A variety of engineered barrier systems can be envisaged, which complement the protection provided by the natural barriers of the geological disposal system. The illustrative geological disposal concept examples considered in this report do not necessarily reflect what the GDF will actually look like for radioactive waste disposal in the UK and we are not committed to any particular geological environment, or to a specific disposal system design. Nevertheless, they help to illustrate why we are confident that we can design, construct and operate the GDF in a range of geological environments in such a way that the long-term safety of people and the environment will be assured.

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# List of acronyms

Acronym	Definition
BFS	Blast Furnace Slag
CSH	Calcium Silicate Hydrate
DNLEU	Depleted, Natural and Low Enriched Uranium
DSSC	Disposal System Safety Case
EBS	Engineered Barrier System
EDZ	Excavation Disturbed Zone
GDF	Geological Disposal Facility
HEU	Highly Enriched Uranium
HLW	High Level Waste
ILW	Intermediate Level Waste
ISA	Isosaccharinic Acid
LLW	Low Level Waste
NAPL	Non-aqueous Phase Liquid
NRVB	Nirex Reference Vault Backfill
OPC	Ordinary Portland Cement
PCM	Plutonium Contaminated Material
PFA	Pulverised Fuel Ash
SF	Spent Fuel
WIPP	Waste Isolation Pilot Plant

# DSSC/452/01

# 1 Introduction

# 1.1 Background

In order to build confidence in the safety of a future geological disposal facility (GDF) for the UK<sup>1</sup>, in the absence of potential disposal sites, RWM is developing a generic Disposal System Safety Case (DSSC), which shows how the waste inventory destined for geological disposal could be safely disposed of in a range of geological environments. Background information on geological disposal in the UK can be found in the Technical Background Document [1].

The documents comprising the generic DSSC are shown in Figure 1 and include a number of research status reports ('knowledge base'). The purpose of the research status reports is to describe the science and technology underpinning geological disposal of UK higher activity wastes by providing a structured review and summary of relevant published scientific literature and discussing its relevance in the UK context. The current suite of research status reports (issue 2) updates and replaces the suite produced in 2010 (issue 1).

Figure 2 shows how research status reports underpin different safety cases. They include:

- reports on the engineered barrier system (EBS) evolution (this report), waste package evolution [2], and geosphere [3], describing the understanding of the evolution of the specific barriers of the multi-barrier system
- reports on behaviour of radionuclides and non-radiological species in groundwater [4] and gas generation and migration [5], describing the release and movement of materials through the multi-barrier system, including the groundwater and any gas phase formed
- reports on criticality safety [6] and on waste package accident performance [7], describing the behaviour of waste packages and a GDF during low probability events
- a report on the biosphere [8], describing how we think the biosphere may evolve in the future and how radionuclide uptake might be expected to take place

Research status reports need to be read in conjunction with other documentation, including:

- the Data Report [9], which describes the values of specific parameters used in the safety assessments based on scientific information presented in the status reports
- the Science and Technology Plan [10], which describes planned future research and development activities

# 1.2 Objectives and scope

The objective of the Engineered Barrier System status report is to summarise the scientific evidence relating to evolution processes of the EBS during periods of storage and after disposal in the GDF. Available information is discussed with the aim of providing a

<sup>&</sup>lt;sup>1</sup> Disposal of higher activity wastes in a GDF is current policy in England, Wales and Northern Ireland. Scottish Government policy is that the long-term management of higher activity waste should be in near-surface facilities. Facilities should be located as near to the sites where the waste is produced as possible.

sufficiently-detailed evaluation of the implications of key processes to allow its direct use in the development of safety cases. Safety-related considerations are excluded from the scope of this document and are provided solely in the safety cases.

The scope covers all materials currently considered in the inventory for disposal, including intermediate and low level waste (ILW/LLW), high level waste (HLW), spent fuels, uranium (particularly depleted, natural and low-enriched uranium, DNLEU) and plutonium.

#### Figure 1 Structure of the generic Disposal System Safety Case (DSSC). The suite of research status reports represents the knowledge base.



# Figure 2 Safety cases and status reports in which underpinning information can be found.

Transport Safety Case		Operational Safety Case		Environmental Safety Case
	Waste P	ackage Evolution Stat	us Report	
			E	ngineered Barrier System Status Report
			Geosphere Status Report	
			Be	ehaviour of Radionuclides and Non-Radiological Species in Groundwater Status Report
			Biosphere Status Report	
		Gas Status Report		
	Critica	ality Safety Status Rep	oort	
Waste Packag	e Accident Performance S	Status Report		
			1	2802-02-N

# 1.3 Audience and users

The primary external audience of the status reports is our regulators. The audience is also expected to include academics, learned societies and stakeholders such as the Committee on Radioactive Waste Management (CoRWM) and Non-Governmental Organisations (NGOs). The reports have been written for an audience with a scientific or technical background and with some knowledge of the context of geological disposal. The primary internal user of the information presented in the status reports is RWM's safety case team.

# **1.4** Relationship with other status reports

There are important interfaces between this and other research status reports. Key interfaces with the Engineered Barrier System status report include:

- the evolution of the waste containers and wasteforms (described in reference [2])
- the evolution of the geosphere, in particular the nature of the groundwater, the timescale for saturation and the nature and magnitude of mechanical loads (described in reference [3]).

Information from the Engineered Barrier System status report that underpins other status reports includes:

- the chemical evolution of the buffer/backfill and the thermal evolution of the EBS, which affect the evolution of the waste packages (described in reference [2]) and the behaviour of radionuclides and non-radiological species in groundwater (described in reference [4])
- impacts of the construction of the engineered barrier system and the materials used on the host rock (described in reference [3]).

# 1.5 Changes from the previous issue

This document updates and replaces the 2010 Near-field Evolution status report [11], published as part of the 2010 generic DSSC suite. Although we have changed the name of the report for this update, we continue to make use of the term near field, which is defined as the engineered barrier system, and those parts of the host rock whose characteristics have been or could be altered by the GDF or its contents. However, detailed discussion of the impacts of the engineered system on the geosphere has been moved to the Geosphere status report [3]. This issue additionally includes understanding from research carried out since 2010, including:

- a cement high temperature study
- an study on enhanced bentonite buffers
- thermal modelling research
- modelling of bentonite saturation
- PhD studies on carbonation of a cementitious backfill
- the Development of Plugs and Seals (DOPAS) GDF sealing project
- results from work on buffer emplacement.

A further change is that while this report describes typical values of parameter ranges to support scientific understanding, the specific parameters used in the safety assessments are presented in a dedicated Data Report [9].

In line with the objectives of the document and in order to respond to previous feedback, contextual and safety-related information have been removed from the text. Contextual

information is provided in [1], while safety-related information is described entirely in the safety case documentation.

# 1.6 Knowledge base reference period

The knowledge base described in this document contains scientific information available to RWM up to March 2016. Where, within RWM's research programme, progress relative to important topics was made after such date, efforts have been made to reflect such progress up to the publication date of this document.

# 1.7 Terminology

For information about use of language and terminology in this and other RWM documents please refer to our Glossary [1]. When necessary, we have introduced specific terminology used in the document through the use of footnotes.

# 1.8 Document structure

The remainder of this report is structured according to the following format:

- section 2 briefly discusses our knowledge base in support of the development of an engineered barrier system for the UK GDF
- section 3 provides a general discussion of the evolution of the engineered barrier system, identifies the main processes that may occur in some example disposal concepts and discusses their potential effects over post-closure time periods
- section 4 provides a summary of the evolution of the engineered barrier system for a range of illustrative geological disposal concepts covering each of the three host rock environments considered in the DSSC
- section 5 presents brief concluding remarks.

We have used coloured boxes at the beginning of each section to provide a short summary of the key messages and help the reader in following the 'golden thread'.

# 2 Development of the engineered barrier system

This section discusses the factors that will need to be considered in developing an appropriate engineered barrier system for the disposal of UK wastes, in support of the more detailed, process level understanding of the evolution of the near field in Section 3 and a summary of the evolution of the engineered barrier system for a range of illustrative geological disposal concepts presented in Section 4.

# 2.1 Selection of engineered barrier systems for the disposal of UK wastes

Meaningful selections of concepts and design components can only be made when we have sufficient site-specific information and understand relevant requirements. A range of materials, both natural and synthetic, are available for fabrication of an engineered barrier system.

Once a specific candidate site (or sites) has been identified, we will gather information on each site's geological environment. This will enable us to identify and assess the possible range of geological disposal concepts, taking account of the waste inventory for disposal, which could be implemented at that site. Examples of illustrative geological disposal concepts that could be developed in different host rocks for different waste types are shown in Figure 3. As this report is concerned with the engineered barrier system situated in the host rock, these examples relate to the host rock only and not to any overlying rocks – overlying rocks are discussed in the Geosphere status report [3]. These illustrative geological disposal concepts are described in the GDF Design Report [12], and are used as a basis for the narratives of near field evolution given in Section 4 of this report.

As our geological disposal implementation programme progresses we will move from highlevel decisions on the concepts to be considered, to concept-specific design decisions on the range of candidate engineered barrier materials and, eventually, to decisions on the designs and materials to be used. The generic knowledge base of the evolution of the engineered barrier system components described in this report will support future decisions. However, we can only make meaningful selections of concepts and design components when we have sufficient site-specific information. Further details on our plans for concept selection are detailed in [13].

# Figure 3 Illustrative geological disposal concept examples for different waste types.

	Illustrative Geological Disposal Concept Examples <sup>d</sup>			
Host rock	ILW/LLW	HLW/SF		
Higher strength rocks <sup>ª</sup>	UK ILW/LLW Concept (NDA, UK)	KBS-3V Concept (SKB, Sweden)		
Lower strength sedimentary $\operatorname{rock}^{\scriptscriptstyle b}$	Opalinus Clay Concept (Nagra, Switzerland)	Opalinus Clay Concept (Nagra, Switzerland)		
Evaporites°	WIPP Bedded Salt Concept (US-DOE, USA)	Gorleben Salt Dome Concept (DBE-Technology, Germany)		

Notes

a. Higher strength rocks – the UK ILW/LLW concept and KBS-3V concept for spent fuel were selected due to availability of information on these concepts for the UK context.

- b. Lower strength sedimentary rocks the Opalinus Clay concept for disposal of long-lived ILW, HLW and spent fuel was selected because a recent OECD Nuclear Energy Agency review regarded the Nagra (Switzerland) assessment of the concept as state of the art with respect to the level of knowledge available. However, it should be noted that there is similarly extensive information available for a concept that has been developed for implementation in Callovo-Oxfordian Clay by Andra (France), and which has also been accorded strong endorsement from international peer review. Although we will use the Opalinus Clay concept as the basis of the illustrative example, we will also draw on information from the Andra programme. In addition, we will draw on information from the Belgian super container concept, based on disposal of HLW and spent fuel in Boom Clay.
- c. Evaporites the concept for the disposal of transuranic wastes (TRU) (long-lived ILW) in a bedded salt host rock at the Waste Isolation Pilot Plant (WIPP) in New Mexico was selected because of the wealth of information available from this United States Environmental Protection Agency (EPA) certified, and operating facility. The concept for disposal of HLW and spent fuel in a salt dome host rock developed by DBE Technology (Germany) was selected due to the level of concept information available.
- d. For planning purposes the illustrative concept for depleted, natural and low enriched uranium is assumed to be same as for ILW/LLW and for plutonium and highly enriched uranium is assumed to the same as for HLW/SF.

The following subsections provide a brief overview of some of the long-term safety factors relating to the host rock that are likely to influence our development of an appropriate engineered barrier system for the disposal of UK higher activity wastes. We also set out the types of materials that might be employed for different engineered barriers.

# 2.2 Technical factors affecting the design of an engineered barrier system

The engineered barrier system developed for application in a particular geological environment needs to be compatible with the nature and quantity of waste, and should complement the barrier provided by the host rock itself.

The nature of the host rock and how this influences the design of an engineered barrier system is discussed specifically for a higher strength rock, a lower strength sedimentary rock and for evaporite rock in Section 4. In any environment, a range of generic characteristics need to be considered in designing an engineered barrier system. These include the following:

• the chemical environment, particularly the mineralogical composition of the local geology and the typical groundwater composition. The presence of potentially

aggressive chemical species, together with the local pH, redox potential and the occurrence of certain microbiological species are all important considerations in evaluating the suitability of potential engineered barrier materials and their compatibility with the natural environment.

- the mechanical stability of the rock, including its strength, flow properties, the location(s) and character of any fault/fracture zones, any differential stresses and the water-bearing properties of the rock. A suitable host rock must be capable of accommodating significant excavation and engineering activities (either with or without engineered supports) throughout the construction and operational phase.
- the thermal properties of the rock, for example, its thermal conductivity. Such considerations affect the required spacing of heat-emitting waste packages and will therefore influence the footprint of the GDF.
- the mechanisms for transport of aggressive, corrosion-inducing species and radioactive substances through the rock. This includes taking account of hydrogeological properties such as the flux of water through the host rock, gas and groundwater transport pathways, the likely extent of advective and diffusive contaminant transport based on the extent of rock fracturing, the nature of the rock matrix, and the presence of sorbing minerals in the host rock.

# 2.3 Selection of engineered barrier system materials

A range of materials are available for fabrication of an engineered barrier system. Appropriate materials will be selected for the buffer, backfill and seals to ensure that the engineered barriers provide complementary functions to those provided by the natural barriers of the specific geological environment.

A range of materials, both natural and synthetic, are available for fabrication of an engineered barrier system. The choice of barrier components and materials to integrate in a disposal system remains open at this stage of the site selection process. Appropriate materials will be selected and combined to ensure that the engineered barriers provide complementary functions to those provided by the natural barriers of the specific geological environment and do not have detrimental impacts on each other [14]. It should be noted that there will also be other materials which do not provide post-closure functions but are present because they have a function in construction, maintenance of the excavations, and/or operations.

Waste packaging options under consideration for UK wastes are discussed further in the Waste Package Evolution status report [2]. Here, a brief overview of the types of materials that might be used for the other engineered barriers, such as buffers, backfills and sealing systems for the GDF, is provided. The selection of a suitable buffer and/or backfill will depend on the functions required for a particular disposal concept.

Typical buffer/backfill materials include [15]:

- clay-based materials such as bentonite. Bentonite is used to refer to smectite-rich material (regardless of origin) with favourable chemical, hydraulic and mechanical properties. Bentonite is the primary buffer and backfill material in a number of HLW and spent fuel disposal concepts. It has also been proposed for use as a buffer and/or backfill in some ILW disposal concepts.
- cementitious materials, which have been proposed as the backfill material in many ILW disposal concepts (as well as an encapsulant/packaging material for ILW). A cementitious buffer is also being actively developed for use in the Belgian 'supercontainer' concept for HLW [16].

- magnesium oxide (MgO), which is used as a buffer material in the Waste Isolation Pilot Plant (WIPP) [17], the underground facility for disposal of US transuranic waste in New Mexico.
- crushed rock, particularly rock that has characteristics and behaviour similar to the host rock. For example, crushed salt is proposed for backfilling the excavated regions of a disposal facility for spent fuel in salt host rocks in Germany. Backfill of this nature might comprise part of the spoil removed during excavation activities.

The choice of materials for engineered seals (for tunnels, shafts and boreholes) within the GDF will depend upon the other materials used as engineered barriers, together with the characteristics of the host rock and the desired longevity of the seals. Designs might incorporate multiple materials, perhaps including:

- bitumen
- clay, including bentonite
- concrete
- crushed rock, or materials closely representing the host rock.

# 3 Generic description of engineered barrier system evolution

In this section a generic description of the main processes that may affect the evolution of the engineered barrier system in some example disposal concepts is discussed, together with their potential effects on the performance of the near field and the host rock. Throughout, we highlight time periods over which these processes occur and explain their likely significance in affecting post-closure safety.

# 3.1 Introduction

Evolution of the engineered barrier system will be an inevitable part of the behaviour of the GDF. We need to understand the processes involved in order to demonstrate post-closure safety.

Disposal system evolution will be an inevitable part of the behaviour of the GDF. In fact, a degree of system evolution of some of the engineered barriers is important. For example, in some disposal concepts it is important for groundwater to saturate the near field and, in doing so, to cause clay-based barriers to swell and develop low permeability. This reduces subsequent groundwater flow and radionuclide migration, while inhibiting microbial propagation (which can lead to the degradation of metallic barriers).

The focus of this report is to give a comprehensive description of those engineered barrier system evolution processes that are likely to be of importance to GDF evolution. Our scenario analysis work [18,19] includes the development and use of internationally accepted procedures (such as feature, event and process (FEP) analysis) to build confidence that all relevant processes have been considered. The extent to which the key near-field evolution processes will occur, and the order and timescales over which they occur, will depend upon the waste inventory, the disposal concept, and the characteristics of the host geology.

At this stage, our aim is to summarise understanding of the processes that might occur. We use our safety assessments to inform and guide future research and development work on the relevant processes, as well as taking due account of stakeholder views on what research should be conducted. Our Science and Technology Plan [10] details the generic (that is, not site-specific) research that we are currently planning in order to address known knowledge gaps, as well as a range of 'curiosity-driven' projects undertaken in collaboration with the Research Councils.

# 3.2 Timeframes for evolution

For the purposes of this report the evolution of the GDF can usefully be subdivided into three timeframes: construction and operations, the early post-closure period and the late post-closure period.

The evolution of a GDF can be subdivided into a number of timeframes that are used in this report and the DSSC Environmental Safety Case [20]. These timeframes are defined rather broadly in terms of events and processes that will occur in the future, rather than in terms of a number of years, since the different characteristics of the illustrative geological disposal concept examples (and the specific characteristics of the host rock and hydrogeology) mean that they will evolve at different rates:

• <u>Construction and operational period, including waste emplacement and closure</u>.

During this period, the GDF will be excavated, drained by pumping (as required), and the facility will be open to the atmosphere. Pumping will affect the hydrogeological system and stress regime around the facility. Ventilation will be used to circulate air throughout the facility. Depending on the method of construction, an excavation disturbed zone (EDZ) will form. Depending on the nature of the local geology, the EDZ may persist or begin to partially or completely reseal. The Geosphere status report [3] contains a comprehensive overview of our understanding of EDZ formation and a narrative of its evolution.

The GDF is likely to be constructed, used for waste disposal and closed over a period of several decades. During this period different areas of the GDF may be at different stages of development. For example at any point in time, new areas of the GDF may be being excavated while elsewhere, in other parts of the GDF, wastes may be being emplaced and engineered barriers installed; this is termed 'phased closure of the GDF'.

• Early post-closure period.

Prior to this period all the waste and the buffer and/or backfill materials will have been emplaced and the GDF will have been sealed. Groundwater will begin to saturate the backfilled excavations and engineered barriers, and hydraulic gradients in both the far-field and near-field rock (similar to those in natural, undisturbed conditions) will be re-established. Depending on the geological environment, this process could develop unevenly in parts of the engineered barrier system and could take hundreds to thousands of years to achieve complete saturation.

Oxygen present in trapped air will be consumed by reactions with the rock and the engineered barriers. The temperature of the near field will begin to rise as a result of heat emission from the waste and exothermic processes involved in the continued maturation of any cementitious backfills. The engineered barriers will begin to evolve in response to various thermo-hydro-mechanical and chemical processes (this will be most significant for HLW and spent fuel disposal areas, however small thermal contributions will also arise from some ILW).

For heat emitting wastes, the early post-closure period is defined by the period during which near-field temperatures peak and then fall back to values close to ambient: effectively the 'thermal period'. During this period waste containers for HLW and spent fuel are designed to remain intact and so will provide complete physical containment of the waste, which is an IAEA requirement [21]. The less corrosion resistant LLW and ILW containers may begin to corrode and will start to release radionuclides into the porewaters through either perforation or through manufactured vents. Radionuclide transport and retardation will then begin. The early post-closure period will range from closure of the facility up to a few hundreds of years to, at most, tens of thousands of years, depending on the disposal concept and geological environment [3].

• Late post-closure period.

The late post-closure period will follow the period in which significant thermal effects may have occurred because of the heat produced by the wastes. The near field will continue to evolve, but the thermo-hydro-mechanical and chemical gradients that may act as the driving force for evolution will be smaller than before. Gradual processes of degradation will affect the engineered barriers, including the waste packages. Eventually, there will come a time when containers for HLW and spent fuel cease to provide complete containment. This may be due to the effects of corrosion, or may arise following a disruptive (geological) event. Nevertheless, the engineered barriers will still continue to contribute to long-term safety by retarding radionuclide migration. The late post-closure period will continue for hundreds of

thousands to a million years after the early post-closure period, depending on the disposal concept and the geological environment [3].

These post-closure timeframes may be quite different for different concepts and geologies. A graphic showing the evolution of a cement based engineered barrier system example is given in subsection 3.10 (Figure 11).

#### 3.3 Key engineered barrier system evolution processes

The key processes that impact on the performance of engineered barriers and/or on how these barriers evolve are best considered in the context of illustrative concept examples.

The processes discussed in this report have been identified as those that may impact on the performance of engineered barriers and/or on how these barriers will evolve over the post-closure period of the GDF. These have been considered in the context of the generic DSSC illustrative concept examples, reflecting our understanding of the relative importance these concepts place on particular processes.

The remainder of this section discusses the main near-field evolution processes that may occur in example disposal concepts and their impacts on the engineered barriers. Section 4 then provides a narrative of our understanding of the evolution of the engineered barrier system in each of our illustrative disposal concept examples (as described in subsection 2.1) for our three illustrative host rock environments.

During the construction and operational period key processes will occur that will need to be considered when developing our site-specific understanding of the evolution of the engineered barriers. These include:

- creation, and partial or complete recovery, of an excavation disturbed zone (EDZ)
- changes in the far field stress regime, including beyond the EDZ
- strong hydraulic gradients during the operational phase
- desaturation and oxidation of part of the host rock immediately surrounding the GDF.

Construction of the GDF will, to some extent, disturb the rock immediately surrounding the excavated space and will create an EDZ. The EDZ will have different mechanical and hydraulic properties to those of the surrounding rock – typically it will be more extensively fractured and more permeable. Depending on the excavation method used, and the nature of the site-specific geology, the EDZ may extend away from the excavated region by a few centimetres to several metres. A detailed discussion of the extent and nature of the EDZ in different geological environments is given in the Geosphere status report [3].

In the early post-closure period, once wastes and the buffer and/or backfill materials have been emplaced and disposal areas sealed, a series of processes will begin that may then continue into the late post-closure period. These include:

- heat generation from the emplaced waste and, to a lesser extent, from exothermic reactions such as those involved in the hydration of cementitious materials, and subsequent thermal effects
- irradiation of engineered barrier materials (particularly of waste package components)
- saturation this evolution of the engineered barriers may include swelling of clay buffers and backfills, and hydration of cement buffers and backfills
- establishment of high pH conditions in cement backfills
- host rock creep (in lower strength sedimentary rocks and evaporites) and compaction of crushed salt buffers

- gradual degradation of engineered barriers, including buffers, backfills and seals, by various processes under lower temperature conditions and in less intense radiation fields compared with the earlier post-closure conditions, leading to long-term evolution of barrier properties and porewater chemistry
- consolidation and evolution of disposal tunnels and shaft seals and plugs
- gas generation due to corrosion, microbial degradation of some organic materials and radiolysis of water and organic materials
- microbial effects
- waste container corrosion in disposal concepts where water is present.

During this period there will be interactions between the wasteforms and near-field porewaters. These are discussed in the Waste Package Evolution status report [2]. Release of radionuclides from the wastes, and radionuclide transport and retardation will also occur during this time. These processes are discussed in the Behaviour of Radionuclides and Non radiological Species in Groundwater status report [4].

# 3.4 Desaturation and oxidation

During the construction and operational period of the GDF, pumping and ventilation will cause desaturation and will result in oxidation of the nearby host rock.

During the construction and operational period, conditions in the GDF will be relatively oxidising as a result of the facility being open to the atmosphere and ventilated. There might also be some inflow of oxygenated near-surface waters and of groundwater. These inflows will be managed through appropriate pumping for the circumstances encountered.

The excavations, together with pumping and processes such as evaporation are likely to alter the natural hydrogeological gradients, causing desaturation, and will also result in oxidation of the host rock around the excavated areas. The spatial extent of these effects will depend on the host rock and the particular conditions encountered, but it is expected that they will be confined to a small region of the host rock around the disposal facility itself.

As soon as the facility is backfilled and closed, the system will start to saturate and, although complete saturation of some parts of the system could take thousands of years, chemically-reducing conditions are likely to be re-established quickly [22]. The reestablishment of saturated, chemically-reducing conditions will halt host-rock oxidation, slow waste container corrosion, and lower the potential solubilities of some radioelements. The effects of the relatively short period of oxidising conditions in the GDF during the construction and operational period need to be taken into account when establishing assumptions regarding the initial conditions for the post-closure safety assessment, and the disposal system must be designed so that desaturation and oxidation do not adversely affect the engineered barriers.

The GDF is likely to be constructed, used for waste disposal and closed over a period of several decades, during which different areas of the GDF may be at different stages of development. Whether the closure of the GDF is phased (that is, some areas may be backfilled and sealed while other areas are being excavated or operated) or takes place as a campaign once all waste is emplaced will depend on a number of factors, including any requirements for the waste to remain retrievable for any period of time [14].

Microbial activity will be present in the open GDF throughout the operational period and could persist for a long period after closure in some regions, depending on the availability of void space, water, nutrients and energy sources. For example, Figure 4 shows a microbial bloom seen at the Äspö Hard Rock Laboratory.



Figure 4 Microbial bloom at the Äspö Hard Rock Laboratory.

Introduced microbes will remain in the GDF indefinitely but, after the early post-closure period, and depending on the concept selected, their contribution to controlling near-field hydrogeochemical processes is expected to diminish. There is uncertainty about their continued role (see, for example, reference [23]) and this is a site-specific factor which will depend upon the overall hydrogeochemical environment of the GDF and its evolution.

The disposal concept and GDF will be designed and constructed so as to minimise potentially detrimental interactions. Where potentially detrimental interactions are unavoidable, the design will take account of their potential effects and magnitudes, for example by using materials that are compatible with each other and by specifying sufficient barrier thicknesses [24].

# 3.5 Heat generation

The thermal evolution of the GDF will depend on a number of factors, including: the rate and amount of heat generated (primarily by radioactive decay and cement hydration), the layout of the GDF, the temperature of the host rock, heat transfer by conduction, convection and radiation, the degree of saturation of the near field and groundwater flow.

During the construction and operational period, and throughout the early post-closure period, a number of processes will generate heat, leading to an increase in the temperature of the engineered barrier system and the surrounding host rock. The most significant heat-generating processes, described in Box 1, are heat produced due to radioactive decay and that due to cement hydration. Other processes, such as corrosion and microbial action (see below) may also generate some heat. However, these processes are unlikely to have a significant effect on the temperature because the amount of heat that will be produced is much less than the heat from radioactive decay or cement hydration. A number of other thermal effects on the engineered barrier system are possible, but are not considered in this report. These include low probability events such as a nuclear criticality after closure or

an accidental fire during the operational phase. These topics are discussed elsewhere [6, 7].

# Box 1 Heat production in the GDF

# Heat produced due to radioactive decay

Heat produced due to radioactive decay is most significant for HLW, spent fuel and, to a lesser extent, plutonium and HEU wasteforms. The IAEA notes that management of decay heat should be considered if the thermal power of waste packages reaches several Watts per cubic metre (possibly less, in the case of wastes containing long lived radionuclides [25].

Heat-generating wastes begin to heat the engineered barriers, the host rock and GDF openings as soon as they are emplaced. The amount of heat produced and the rate of heat production will depend on a range of factors that determine the radionuclide inventory in the waste, such as the processes that led to waste generation and the duration of interim storage prior to disposal. Radiogenic heating from spent fuel is expected to be significant for a few thousand years<sup>2</sup> after disposal, while short-lived radionuclides decay [14]. Contributions will also arise for Pu wastes (contributions from <sup>238</sup>Pu, <sup>239</sup>Pu, <sup>240</sup>Pu and the ingrowth of <sup>241</sup>Am daughters).

# Heat produced due to cement hydration

The hydration of cementitious materials proceeds via a range of exothermic (heatreleasing) chemical reactions. Some of the hydration reactions occur rapidly, releasing significant quantities of heat quickly and can cause a significant temperature increase (persisting for a few years to several tens of years<sup>3</sup>). Therefore, the temperature of a GDF engineered barrier system that includes large quantities of cementitious materials, for example, as backfill in disposal concepts for LLW and ILW, may be raised as a result of cement hydration. Various research studies for such disposal concepts indicate that near field temperatures may be elevated by several tens of degrees for a few decades [22, 26]. The heating that may occur due to cement hydration is less than the heat produced due to radioactive decay of HLW and spent fuel.

The maximum temperatures that may be reached in the engineered barrier system and the thermal evolution of the GDF will depend on a range of factors, including [27]:

- the quantity of heat generated and the rate of heat generation
- the temperature of the surrounding host rock (the geothermal gradient increases with depth, as discussed in the Geosphere status report [3] this is particularly significant for high heat generating wastes)
- the layout of the GDF (particularly the spacing of heat-generating waste packages and the size and arrangement of any vaults to be backfilled with cementitious materials)
- heat transfer by conduction and the thermal conductivities of the host rock and engineered barriers
- the degree of saturation of the near field
- heat transfer by convection.

<sup>&</sup>lt;sup>2</sup> Example timeframe provided, based on thermal modelling carried out by Nagra and SKB [28,48]. RWM is currently undertaking thermal modelling to determine the thermal heat output associated with the disposal of UK wastes (see Tasks 456-460 in our Science and Technology Plan [10])..

<sup>&</sup>lt;sup>3</sup> This depends partly on the mass of material involved.

During the operational period, ventilation will remove heat produced in the disposal facility. Once the GDF is closed, the temperature will rise, reach a maximum value and then slowly fall, as heat is dissipated by conduction, convection and radiation processes and as heatgenerating processes diminish.

For the post-closure phase, the Disposal System Technical Specification document [14] currently specifies maximum temperature targets for different disposal areas and waste types. For ILW and LLW disposal areas, a guidance value of less than 50°C for all waste packages following closure is given [14]. The consequences during any period when this target is exceeded (for example, in response to backfill curing should a cementitious backfill be used) are also considered. In UK ILW disposal designs that incorporate a cementitious backfill, waste package temperatures of up to 80°C are allowed for a period of 5 years.

For HLW and spent fuel disposal areas in higher strength rock a maximum peak temperature of 100°C on the external surface of the disposal container is currently given in order to prevent thermal processes from impairing the mechanical properties of the container material and the properties of the buffer [14, 28]. For their KBS-3V concept, SKB recommend that the buffer temperature should not exceed 100°C in order to limit mineralogical alteration of the bentonite [29, 30]. However, higher thermal limits could also be specified on the basis of the design selected [14] depending on how stringent the requirements on the buffer are.

The disposal of high heat generating wastes in a GDF creates a number of technical questions that need to be addressed in order that a safe disposal solution can be developed. RWM established a high heat generating waste project specifically to address these questions [31]. The aim of this project was to enhance the understanding of the factors affecting geological disposal of high heat generating wastes with a view to supporting the development of the disposal system specification for these wastes (capturing the disposal system requirements) and spent fuel life cycle options (for example, in support of the development of packaging solutions). This project included the development of a Thermal Dimensioning Tool (TDT) [32] that:

- uses analytical and semi-analytical expressions to solve the heat conduction problem to take full advantage of speed and accuracy inherent in these approximations, when allied to simple geometrical configurations of the waste
- has the ability to perform thermal dimensioning<sup>4</sup> for a range of possible disposal concepts for high heat generating wastes
- can model the consequences of parametric uncertainty
- supports good principles of quality assurance of data
- has a simple, clear user interface to help the user construct a model
- produces graphical output to show the evolution of the temperature with time.

The TDT is therefore able to perform thermal dimensioning analyses in order to quantify the consequences of potential temperature constraints on the engineered barrier system (such as that the temperature of the buffer material is within specified limits).

<sup>&</sup>lt;sup>4</sup> Thermal dimensioning refers to thermal modelling aiding the design and sizing of a GDF.

# 3.5.1 Modelling temperature elevation in GDF disposal areas

Thermal modelling is carried out to understand the thermal evolution of different GDF concepts and designs; it can be used to underpin the selection of temperature limits.

To underpin the selection of thermal limits for specified geological disposal facilities, thermal modelling has been conducted for a wide range of different disposal concept designs [26, 33, 34, 35, 36, 37].

For HLW and spent fuel disposal concepts, thermal modelling can be used to calculate the temperatures to which engineered barriers would be exposed [33]. In particular, for concepts that employ a bentonite buffer, such modelling activities are used to inform design decisions and waste acceptance criteria with regard to the high-temperature dependence of bentonite performance (see 3.5.2).

Figure 5 illustrates an example of thermal modelling reported by Nagra applicable to their concept for the disposal of spent fuel in Opalinus Clay [33, 38]. It can be seen that beyond the disposal container surface the temperature in the outer half of the bentonite after 100 years is calculated to be below 100°C. The input parameters used in the example of thermal modelling illustrated in Figure 5 are detailed in reference [38].





More recent thermal modelling work conducted by Nagra considered thermal evolution at the container surface, taking into consideration relative humidity and corrosion. This indicates that considerably lower temperatures would occur in a disposal area for spent fuel in Opalinus Clay [39]. In these calculations the temperature at the container surface would increase to a peak surface temperature of 120°C to 140°C, achieved after about 5 to 10 years after waste emplacement. Thereafter, a long, slow cool-down period is calculated, together with descriptions of the expected corrosion products and processes occurring at the container surface at such timeframes (see reference [39]). These temperatures are consistent with the approach adopted by Nagra to ensure that the outer half of the bentonite buffer remains unaffected by thermal alteration processes.

The temperature limit on the surface of the container or buffer will depend on the requirements for the engineered barrier system selected. In some cases a specific value might not be required<sup>5</sup>, for example, when utilising a sufficient thickness of bentonite that the buffer performance is retained in the cooler outer region, even if the buffer in direct contact with the container is affected by thermal alteration

Some of the justifications given by waste disposal agencies for the temperature limit specifically mention the transformation of smectite into illite. A recent study [40] has indicated that such illitisation is unlikely to result in sufficient deleterious effects to a bentonite buffer. The study did however note that the potential for steam to result in loss of bentonite swelling pressure warrants further consideration. The effect of steam on bentonite has also been looked at in another study [41], which noted large differences in final volume after active dispersion between reference and steam exposed material, however no evidence of short-term exposures to steam impairing the swelling properties of bentonite were found.

Another study, as part of the high heat generating waste integrated project [31], has considered enhancing the thermal conductivity of the buffer material immediately surrounding the container as one means of managing the temperature of the buffer [42]. One approach considered is to mix the bentonite with materials of a higher thermal conductivity, such as sand or graphite. The study concluded that comparatively modest levels of thermal conductivity enhancement arising from the addition of sand offered little benefit, whereas the addition of graphite shows greater promise. However, there is significant uncertainty concerning how all aspects of the buffer would be affected by the addition of graphite, since graphite can also enhance container corrosion.

A research project to understand the post-closure thermal impact of HLW and spent fuel waste packages has recently been completed [43]. This study included a simple qualitative analysis of potentially relevant processes to identify those processes, and couplings between processes, that should be considered during the post-closure thermal evolution of a disposal facility for high level waste and spent fuel. A stepwise modelling approach was used, beginning with simple baseline calculations that considered only thermal conduction and radiation and progressing to fully coupled thermal-hydraulic calculations. The conclusions were that:

- the thermal density of the source term (determined by the combination of waste type, package loading, cooling time and package spacing) is the key control on the temperatures seen in the immediate vicinity of the waste package
- the choice of materials (host rock geology, buffer material) has a lesser influence
- thermal-hydraulic coupling via the material properties can significantly influence thermal evolution
- drying of the buffer immediately adjacent to the waste package results in a decrease in thermal conductivity compared to the as-emplaced properties, which leads to an increase in the temperature of the buffer close to the waste package.

The evolution of bentonite buffers more generally is the subject of subsection 3.9.

Thermal modelling has also been undertaken for ILW/LLW disposal areas. The most recent work undertaken for the UK ILW/LLW illustrative concept for a higher strength host rock includes input data such as radionuclide decay and the calculated cement backfill exotherm [44]. Such modelling indicates that peak temperatures in the disposal area within a 50 year

<sup>&</sup>lt;sup>5</sup> Nagra do not currently have a temperature limit for the buffer for the Opalinus Clay disposal concept.

transient after backfilling would be no greater than 40°C<sup>6</sup> (covering the wasteform, the container and the backfill), although this depends on assumptions about the ambient temperature at the depth of the facility – the reference case considered an initial condition of 25°C. Ventilation during the operational phase (over several decades) strongly influences the temperature distribution in the local host rock. If ventilation is maintained during backfilling it would draw a significant part of the backfill exotherm heat loading out of the facility. If the temperature of the ventilation system was increased or ventilation was stopped during backfilling then the local temperatures in the packages may be increased.

# 3.5.2 Impacts of elevated temperature on the evolution of the engineered barrier system

Many of the processes occurring in the engineered barrier system will be strongly coupled to temperature. Elevated temperatures will affect the chemistry of the engineered barriers, and will cause thermal expansion of engineered barrier materials and the host rock. Due to faster reaction kinetics at elevated temperatures, cementitious materials tend to become more crystalline. For clays, thermally-driven mineral alteration processes may lead to the development of new mineral phases.

Many of the processes occurring in the engineered barrier system will be strongly coupled to temperature. Consequently, heat generation may cause a range of effects that influence the evolution of the near field and affect the performance of the engineered barriers [45].

Elevated temperatures will affect the chemical evolution of the engineered barriers, altering thermodynamic equilibria and tending to increase the kinetics of reactions [30]. A temperature rise will cause thermal expansion of engineered barrier system materials, the host rock and near-field porewater, and could induce mechanical stresses. Such stresses may dissipate gradually with cooling, but may also cause cracking in some materials [46, 47, 48, 49], see subsection 3.10.2.

The influence of temperature on the performance of the waste package and on gas generation is described in the Waste Package Evolution status report [2] and the Gas status report [5] respectively. In general, higher temperatures tend to increase the rate of corrosion processes and associated gas generation rates. However, by the selection of appropriate container materials, together with the selection of other engineered barriers (such as buffers and backfills) and appropriate controls on the thermal evolution of the GDF, waste packages can be designed to provide the required functionality over a specified time period. For example, for the illustrative higher strength host rock HLW and spent fuel disposal concept, where temperatures are not expected to exceed 100°C [29], disposed waste packages can be expected to provide complete containment for potentially 100.000 years or more [50, 51]. Another example is the UK illustrative ILW/LLW disposal concept [22], where increased temperatures may lead to higher gas generation rates and possible over-pressurisation within the waste package. Here, over-pressurisation is prevented through a combination of appropriate container design, such as the use of vents which prevent unacceptable build-up of internal gas pressure. A high porosity and high permeability backfill (relative to the host rock) facilitates gas migration and prevents excess pressurisation in the engineered barrier system.

<sup>&</sup>lt;sup>6</sup> Although deviations from the reference case conditions may lead to short-lived excursions above this value.

#### Cement-based buffers and backfills

For cement-based buffers and backfills, elevated temperatures will impact on the evolution of these barriers through a variety of alteration mechanisms which have been extensively investigated (for example, see reference [26]). Due to faster reaction kinetics at elevated temperatures, cementitious materials tend to become more crystalline with time, particularly if the materials are exposed to elevated temperatures for an extended period [22]. Cementitious materials exposed to elevated temperatures would have a lower pH buffering capacity but would be able to provide such a buffering capacity (the ability of a buffer to withstand change in chemical conditions) for longer. This is caused by the lower solubility of the calcium hydroxide  $(Ca(OH)_2)$  constituent of the cement and the increased dissociation of water at higher temperatures [52].

The degree of thermal alteration to be expected is uncertain and will depend on the composition of the cement material used, together with the thermal evolution of the disposal area following backfilling. Ageing and alteration mechanisms due to elevated temperatures will modify properties of the cement backfill in respect of pH buffering performance and surface area (a factor influencing radionuclide sorption). However, experimental evidence and comparison with natural analogue minerals [53] show that hydrothermally-altered cement phases will continue to provide the required porewater pH buffer capacity for a specified time. For example, for the UK ILW/LLW illustrative disposal concept it is expected that the pH in a cementitious engineered barrier system will be maintained at more than pH 10 for at least one million years, even when taking into consideration such thermal effects [54].

A review of cement performance at high temperatures (in excess of 100°C) [55], as part of our high heat generating waste project, concluded that the effect of exposing cementitious materials to such elevated temperatures, whether or not the temperature is sustained for a long period, is to cause a reduction in strength and stiffness. However, at a maximum temperature of 300°C, assumed for the review, these changes are not so significant that they could not be dealt with in the design process. Effects of temperature on cementitious materials at more moderate temperatures are the subject of a recent review of the types and properties of cementitious materials [56, Section 7].

# Clay-based buffers

Where clay-based buffers<sup>7</sup> are considered for use in HLW and spent fuel disposal concepts (see, for example, references [33, 29]), elevated buffer temperatures (in the range of 100 to 150°C, depending on the disposal concept [27]) may be reached due to the heat generated by these wastes. It should be noted that after resaturation, the pressure at GDF depth will mean the boiling point of water will be significantly elevated. For such systems, drying of the clay material may occur if water can be transported away from the buffer, which is unlikely and a deviation from planned conditions. This may extend the time required for the buffer/backfill to become saturated, or may cause desaturation of the buffer if significant water is lost and the swelling of minerals in the bentonite reduced. Installed buffer blocks will almost certainly crack from elevated temperature/temperature gradients [57], see Figure 6, but this is not an obvious issue since there are already joints between blocks and these are expected to disappear as the bentonite wets and swells. The extent to which heat generation causes drying will also depend upon other factors such as the rate of groundwater ingress from the surrounding host rock. There is also the potential

<sup>&</sup>lt;sup>7</sup> Buffer here refers to clay-based barriers in typical HLW and/or spent fuel disposal concepts that protect the waste package and limit migration of radionuclides following the eventual failure of the waste container.

complicating factor of the dissociation of water due to the anaerobic corrosion of steel in those concepts with a steel waste package

# Figure 6 Cracks observed from the inside of ring-shaped bentonite blocks (from a full scale test carried out by SKB [57]).



Exposure to elevated temperatures may cause mineralogical alteration in a clay-based buffer or backfill [30]. Upon saturation of bentonite buffers used as engineered barriers in HLW and spent fuel disposal concepts, water will be taken up by the minerals in the bentonite which will cause the buffer to expand and swell. The continued swelling of the bentonite buffer and backfill will be resisted by the rock walls of the GDF and a swelling pressure will develop, creating a low permeability barrier around the waste packages that inhibits advective flow. Thermally-driven mineral alteration processes may lead to the development of new mineral phases with degraded performance. Such processes include illitisation, as described in Box 2. There is some uncertainty over the temperature at which potentially detrimental mineral transformations occur in clay-based buffers. Some evidence suggests that the performance of typical clay-based buffer materials, such as bentonite, may be affected by temperatures as low as 100°C [27], whereas other sources indicate that mineralogical transformations relevant to GDF timeframes may not occur until much higher temperatures [58].

In general, thermally-induced processes have the potential to impact the performance of materials used for buffers and/or backfills for HLW and spent fuel disposal concepts more than those used in ILW and LLW disposal concepts. To ensure that the impacts of increased temperatures are properly managed, temperature limits will be imposed on the disposal system for all disposal areas so as to preclude any significant thermal effects relevant to performance [27].

# Box 2 Illitisation – transformation of smectite to a non-swelling clay

Smectite, a swelling clay mineral, is the main constituent of bentonite and dominates its physical and chemical behaviour [58]. When exposed to higher temperatures and pressures, smectites are unstable and are transformed to more stable silicate phases, such as illites. This process may lead to a significant change in swelling pressure and hydraulic conductivity of the clay. Typical swelling pressure specifications for swelling clays are of the order of 1 MPa [59]. The plot below, taken from reference [59], illustrates how temperature can influence the percentage of illite in a range of naturally-occurring smectite-containing clays sampled from different sedimentary basins.



Illite content of natural clays exposed to a range of temperatures

The transformation of smectite to illite is strongly controlled by kinetic and hydrological constraints, such as:

- the time of exposure to enhanced temperature and pressure
- the concentration of exchangeable cations, for example potassium ions
- the saturation state in the bentonite
- the permeability of the local host rock (adjacent formation).

However, it should be recognised that other reactions (for example zeolitisation) may be equally as important as the smectite to illite reaction. Potential reactions will need to be assessed via an assessment of mass balance, mass action and kinetic constraints [60].

The effects of mineralogical alteration and associated local volume and stress changes, potentially leading to porosity reduction and/or cracking, can be factored into estimates of uncertainty in near-field permeability and radionuclide transport. Work is on-going within our research programme to support the development of a well justified thermal limit for a UK GDF for a variety of potential disposal concepts (see Tasks 456-460 in our Science and

Technology Plan [10]). Further work is also required to improve our understanding relating to the extent of alteration of clay-based materials when exposed to temperatures in the 100°C to 150°C range and how any clay alteration (and/or drying) would impact on saturation processes (see subsection 3.7.1 and Tasks 464 and 472 in our Science and Technology Plan [10]).

# 3.5.3 Coupled thermal effects

Coupled thermal processes may indirectly impact on the ability of an engineered barrier to perform the required function(s) and therefore thermo-mechanical and thermo-hydro-mechanical effects need to be understood.

In addition to direct thermal impacts, as discussed above, there are a number of coupled thermal processes that may also indirectly impact on the ability of an engineered barrier to perform its required function(s) over the desired timeframe. Thermo-mechanical and thermo-hydro-mechanical effects therefore may need to be evaluated, particularly for those parts of the engineered barrier system where temperature gradients are highest. Interfaces between different materials that may have different thermal properties, and the potential for local thermal gradients, may also need to be considered. Some important coupled thermal effects are listed below:

- increased temperatures will tend to enhance the activity of microbial populations naturally present in the host rock, as well as those introduced during GDF construction and operations. This may accelerate corrosion (for example, pitting corrosion) and gas generation in the near field. At higher temperatures, some microbial populations may be less capable of activity or survival, but new microbe populations could develop [61, 62]. There is uncertainty over the impact of such processes, and this may require further investigation in the future.
- a high temperature gradient in bentonite maintains low moisture levels in the hottest part of the bentonite, even when saturation of the bentonite is approached. This results in a low moisture level at the container surface for many years, helping to reduce corrosion of the container [33].
- increased temperatures will lead to thermal expansion of porewater and reduced viscosity of this fluid, therefore affecting clay buffer pressurisation. This will help to increase the porewater pressure and swelling of the bentonite, providing the local access tunnels are backfilled and sealed, which would have a positive effect on its performance.
- in evaporites, higher temperatures will increase the convergence rate due to creep of the crushed salt backfill. The extent of such a process will depend on the heat production characteristics of the wastes emplaced and the disposal area design.
- in evaporites, heating may also promote the migration of brine pockets and other fluid inclusions in the salt towards the GDF along a thermal gradient, which could cause water to accumulate in the near field and potentially enhance container degradation rates [63].
- elevated temperatures are likely to lead to stresses caused by the thermal expansion of engineered barrier materials. However, during the temperature decreasing phase, thermal contraction may reduce compressive stresses or even induce tensile stresses, which may cause cracks in these materials. This effect is discussed further with respect to cracking of cement backfills and seals in subsection 3.10.2.
- cracking of cementitious backfills due to thermally-induced wasteform/waste package expansion [49] and corrosive expansion of wasteforms [64].

- elevated temperatures may affect saturation rates indirectly as a result of gas generation, by altering pathways for fluid migration, by accelerating fluid migration rates and/or by increasing the pressure in the GDF<sup>8</sup> [65]. In some cases reducing the rate or extent of saturation may affect the properties of engineered barriers by causing shrinkage and/or cracking. Again, these uncertainties may need to be considered in the forward programme of research, depending on the nature of the geology and disposal concepts taken forward.
- impacts of a thermal gradient arising from one different disposal area on another disposal area also need to be considered. For example, designs will need to consider appropriate separation of disposal areas that may attain higher temperatures (for example, spent fuel) from other areas (such as those containing cement) [66] to ensure different areas do not have a significant effect on each other [67] – see the Geosphere status report [3].

#### 3.6 Ionising radiation

lonising radiation is emitted by radioactive substances as they decay. Radiation will interact with all components of the wastes, with the engineered barriers, and with the near-field porewater.

lonising radiation is emitted by radioactive substances as they decay. Depending on the origin of the radioactive wastes under consideration, the level of radiation that is emitted will vary. The expected levels of radiation at the wasteform surfaces will be significantly greater for HLW and spent fuel compared to ILW/LLW.

The direct effects of radiation emitted by the radioactive waste will diminish with increasing distance from the waste, as each additional barrier provides a successive layer of shielding. Thus, the radiation levels will be most intense within the wasteform, somewhat lower for the container, and lower still in the buffer, backfill and host rock.

Radiation will interact with all components of the wastes, with the engineered barriers (the waste matrix, the waste container and backfill), and with the near-field porewater. This could result in effects on corrosion, radionuclide speciation and the degradation of near-field components, including the wastes. These processes may be affected by the direct interaction of ionising radiation with materials (such as solid organic polymers and water), reactions with the radiolysis products of water, or through alteration of pH or oxidation potential due to the effects of radiolysis (these processes may also be inter-related). The radiation dose will differ at different places within the GDF due to the different radionuclide contents of waste containers arising from different waste streams.

#### 3.6.1 Radiation damage to engineered barrier system components

lonising radiation generated by radioactive waste will interact with the engineered barrier system materials to produce chemically-excited species, some of which may promote metal corrosion.

The impacts of ionising radiation on waste package and disposal container performance are described in the Waste Package Evolution status report [2]. The ionising radiation generated by radioactive waste will interact with the materials inside and surrounding the waste package to produce chemically-excited species. Some of these species will tend to promote metal corrosion (they are oxidants). For ILW packages and thick-walled HLW and

<sup>&</sup>lt;sup>8</sup> Pressurisation by increased gas generation could delay full resaturation in some disposal concepts.

spent fuel disposal containers, the potential for radiation-assisted corrosion is low due to the relatively low radiation dose rate at the containers' external surfaces.

Materials considered for use as wasteforms for HLW and spent fuel, such as borosilicate glass and ceramics (including spent fuel) are typically highly resistant to radiation damage. Similarly, encapsulants used for ILW/LLW are also tolerant to radiation damage. Radiolysis (splitting) of water molecules generates reactive chemical species such as hydroxide radicals, as well as molecular hydrogen and oxygen. Over the long term, radiation may cause processes such as cracking, swelling or recrystallisation of a wasteform matrix, which may enhance the potential for release of radionuclides from the wasteform into porewaters. Consideration of the impacts of radiation on the wasteform is described further in the Waste Package Evolution status report [2].

With regards to the impacts of radiation on the performance of the waste container, both modelling and experimental work have been undertaken to investigate the rate of gas generation in the presence of representative radiation fields, to analyse the composition of the corrosion products formed from candidate container materials, and to determine whether radiation has an effect on anaerobic corrosion behaviour [2, 68]. Using such information, the impacts of radiation damage can be factored into calculated corrosion rates for candidate container materials used in post-closure safety assessments [2]. Radioactive gases, generated as a result of radiolysis of waste and of organic materials (see subsection 3.6.2), is included in the gas source term used in post-closure safety assessments and the Gas status report gives further information [5].

For HLW and spent fuel engineered barrier systems that employ a clay-based buffer, the radiation penetrating through the thick waste container to the buffer is highly attenuated. For Swedish spent fuel disposal, the maximum dose outside the container is expected to be no more than 500 mGy per hour [30], and will be predominantly due to the decay of relatively short-lived radionuclides, so this will not be significant for prolonged periods (not beyond 1000 years) [29]. The majority of gamma radiation from the waste is shielded by the iron and copper in the container, so only a minor fraction ever reaches the buffer while the container remains intact (a period in excess of 100,000 years for this design of container).

A number of investigators have studied the extent of enhanced decomposition of montmorillonite, a mineral in bentonite that contributes to its swelling capacity, when exposed to radiation [69, 70]. More detail on the chemical composition of bentonite is given in subsection 3.7.1. The results of these investigations suggest that the accumulated dose in the buffer will be too low to affect its ability to swell and thereby the ability of the buffer to control the rate at which groundwaters can move to (or from) the waste container will be maintained. Similar conclusions can be reached for the effects of radiation damage on the backfill (since the radiation field is even lower in these regions) [69].

For HLW and spent fuel engineered barrier systems, once the waste container has been breached, radionuclides may migrate out of the wasteform into the near field. From this point, additional radiation damage processes may be possible, such as from alpha decay of radionuclides sorbed on the bentonite. However, only a small part of the buffer would be affected [30] and only a small fraction of the inventory would remain. Only if early waste container penetration were to occur, would small parts of the buffer be affected by exposure to higher dose rates. In this event, several clay mineral properties such as solubility, specific surface area, and ion-exchange capacity could be altered by local damage produced by radiation, but the effects on properties appear significant only for high doses and remain relatively limited in spatial extent [70].

For disposal systems that employ a cement-based backfill, typically ILW/LLW disposal areas, exposure to radiation will, in general, be much less than compared with a HLW and spent fuel disposal area and so no significant impact on the performance of cement-based backfills is expected. Cementitious materials considered as backfills (similar to
cementitious wasteforms) in general have good resistance to physical degradation when exposed to radiation although, as discussed in the Waste Package Evolution status report [2], some wasteforms associated with specific wastes (such as those containing large proportions of organic material) may be less resistant [22]. The most important impact considered with respect to cement-based disposal concepts is how radiolysis of water could affect the redox potential of near-field porewater; this is discussed further in subsection 3.6.2.

#### 3.6.2 Impacts of radiolysis on near-field redox conditions

Oxygen generated from water radiolysis may cause localised oxidising conditions to persist for longer. In general, however, the effects of radiation damage and radiolysis outside the waste container are negligible.

Oxygen generated from water radiolysis will tend to dissolve in the near-field porewaters and may consume reducing species, for example ferrous (Fe<sup>2+</sup>) cations, and hence cause localised oxidising conditions to persist for longer, particularly in areas of the engineered barrier system that are exposed to a high radiation field.

In general, impacts of higher redox potential as a result of radiolysis will be most significant for engineered barrier components exposed to high levels of radiation. High level wasteforms are dry and so are only affected by increased redox potential as a result of radiolysis should early container failure occur. Even in such an event, higher redox potential is unlikely to impact the container through increased corrosion rates (see the Waste Package Evolution status report [2] for further discussion). However, this could be the case for spent fuel, the dissolution behaviour of which may be redox-sensitive. Additionally, for spent fuel, water associated with the fuel rods (from the storage of spent fuel elements in water 'ponds' during cooling) will largely be removed through drying prior to packaging in waste containers; however a small residual amount of water may still remain. For example, SKB and Posiva specify that the amount of residual water associated with the fuel rods in a single disposal container should be no greater than 600 grams [30, 711<sup>9</sup>. Since radiation levels inside a spent fuel waste container will be relatively high. especially during the early post-closure period, radiolysis of water present could result in pressurisation and internal corrosion. This is further discussed in the Waste Package Evolution status report [2].

Once a spent fuel waste container is breached, groundwater could enter the container and waste dissolution would begin. Changes to redox potential due to radiolysis of water can impact on the rate of wasteform dissolution (see the Waste Package Evolution status report [2]) and could impact on the solubility of redox-sensitive radionuclides once released from the wasteform (see Behaviour of Radionuclides and Non radiological Species in Groundwater status report [4]).

A number of investigators have also studied the radiolysis of porewater [30], which could affect the chemistry in the buffer (for example, the redox potential). In general, investigators have concluded that radiolysis of buffer pore water will be insignificant, as the dose rate outside the waste container will be too low to have any effect [30]. Similar conclusions were reached for the effects of radiolysis in the backfill (as the radiation field is even lower in these regions).

For cement-based ILW concepts, the contents of some waste packages will be more radioactive than others, and water radiolysis may cause oxidising conditions to persist for

<sup>&</sup>lt;sup>9</sup> Illustrative value – the maximum permissible quantity of water inside a Swedish sealed container is set at 600 g, corresponding to 12 leaking fuel rods filled with water, assuming that the void space inside one fuel rod is 50 cm<sup>3</sup>.

longer in these packages than in those where the radiation field is less intense. The possible impact of radiolysis on the evolution of an ILW/LLW disposal area is discussed more fully in reference [72].

In general, radiation fields in geological disposal facilities are typically such that the amount of radiation damage is very low, and the effects of radiation damage and radiolysis outside the waste container are negligible [73].

# 3.7 Resaturation of the near field

Once the GDF is closed and sealed, groundwater will enter the EBS and the facility will begin to resaturate. The timescale for resaturation will depend on: desaturation during operations, hydrogeological properties of the geosphere, local spatial variability, permeability of materials, thermal evolution, any gas generated and the size of the GDF.

During the construction and operational period, inflow of water into the disposal facility may begin to occur. The extent of resaturation<sup>10</sup> during this period will vary considerably, depending principally on the disposal concept, the host rock, the engineered barrier materials used and the local hydrogeology. It may be necessary to implement a range of water inflow management measures during the operational period. Such measures may include the sealing and/or avoidance of locations where there are particularly active flowing fractures. The effects of water inflow during resaturation will need to be considered in the site-specific post-closure safety case.

In general we expect that resaturation of the engineered barrier system will begin predominately during the early post-closure period once the facility is closed and sealed. During this time groundwater will enter the near field and the facility will begin to resaturate. Illustrative timescales of resaturation for various disposal concepts are described in Box 3. The timescale for resaturation is dependent on a range of factors, including:

- the degree and spatial extent of de-saturation that had occurred during the construction and operational period
- the large-scale hydrogeological properties of the geological environment, including hydraulic gradients and the distribution of hydraulic conductivities that control the rate of groundwater ingress into the near field
- the porosity of the host rock
- the mechanism(s) and local variability of groundwater ingress to the engineered barriers (for example, whether advection or diffusion dominates and whether uniform or channelled flow occurs)
- the role of any suction processes (such as the thermally-induced migration of porewater into the facility)
- the permeability of near-field materials
- the occurrence of gas generation and gas flow
- the size and geometry of the GDF.

<sup>&</sup>lt;sup>10</sup> Strictly speaking, this is the first time the material is saturated, but resaturation is the accepted term for this process – meaning resaturation of the excavation rather than the material.

# Box 3 Predictions of engineered barrier system resaturation

Reaturation timescales for materials used in different disposal areas of the GDF. The rate of resaturation may also be spatially variable, depending on local hydraulic properties of the near-field rock.

For example, for disposal concepts employing a bentonite buffer in higher strength host rock, typical estimates using multi-phase flow process models estimate complete resaturation times ranging from tens of years to thousands of years, as a result of spatial variability of flow in the fracture network [74]. Predictions for resaturation in a lower strength sedimentary host rock range from hundreds to tens of thousands of years, reflecting the degree of uncertainty regarding this process and the range of values and parameters that control it [33].

For cement-based concepts, calculations of resaturation rates using both analytical calculations and coupled thermo-hydraulic calculations incorporating two-phase flow (which include taking into consideration the impact of gas generation on resaturation times of the near-field), give estimated resaturation times for ILW emplacement tunnels of the order of hundreds of years in a lower strength sedimentary host rock (the maximum hydraulic conductivity of Opalinus Clay is 10<sup>-13</sup> m s<sup>-1</sup>) [33]. In comparison, estimates for disposal area resaturation for a higher strength host rock could be of the order of tens of years to a few hundred years [22].

Resaturation is a complex process that is still relatively poorly understood, especially in low-permeability materials such as those that are likely to be used in the near field of the GDF. Resaturation will not occur uniformly throughout the facility because of variations in the properties of the engineered barrier system components and heterogeneities in the host rock. For example, in addition to variability in near-field rock hydraulic properties, buffer resaturation rates are also affected by temperature, swelling pressure, geometry, buffer mineralogy and groundwater composition. We do not yet fully understand the degree to which such heterogeneities will influence the performance of near-field barriers, either before or after they become resaturated. However, we are involved in the planning of future research on bentonite heterogeneity (see Tasks 466 and 473 in our Science and Technology Plan [10]). Resaturation functions, as discussed in subsection 3.7.1. It is also through the process of resaturation that a cement-based backfill provides a chemical containment function by buffering near-field porewaters to high pH, as discussed in subsection 3.10.1.

# 3.7.1 Resaturation of bentonite buffers

Resaturation of bentonite buffers used as engineered barriers in HLW and spent fuel disposal concepts is important because as water is taken up by the minerals in the bentonite, a swelling pressure will develop to create a low permeability barrier. There is however considerable uncertainty regarding the timescale for complete resaturation.

Resaturation of bentonite buffers used as engineered barriers in HLW and spent fuel disposal concepts will begin during the construction and operational period and will continue in the early post-closure period. The rate of resaturation will depend on the local hydrogeological conditions of the host rock. As noted in subsection 3.5.2, once groundwaters contact the bentonite buffer (and backfill) water will be taken up by the minerals in the bentonite (Box 4 describes the minerals in bentonite), which will cause the buffer to expand and swell; a swelling pressure will develop, creating an impermeable barrier around the waste packages that inhibits advective flow.

# Box 4 Bentonite mineralogy

The term bentonite is used to refer to smectite-rich material regardless of origin. The bentonite that is typically specified in engineered barrier systems consists primarily of calcium or sodium exchanged montmorillonite (a smectite clay mineral), with minor amounts of quartz, feldspar, kaolinite, carbonates, sulphides, sulphates and organic matter [75].

#### Mineral compositions (wt%) of example bentonites (sub-set taken from [27]).

Mineral	Bentonite			
	MX-80	Deponit CA-N	FEBEX	S-2
Notes [27]	Reference bentonite buffer material being considered in Sweden, Switzerland and other countries – a natural sodium bentonite from a deposit in Wyoming, USA.	An alternative potential buffer material considered in Sweden - a natural calcium bentonite from a deposit on the island of Milos, Greece.	Used by Enresa as buffer material in FEBEX project – from the Cortijo de Archidona deposit in Spain.	Tested as a reference buffer by Enresa – also from the Cortijo de Archidona deposit.
Montmorillonite	87	81.4	89-95*	88-96*
Quartz / Chalcedony	3	0.4	1-3	1-3
Cristobalite	2	0.6	1-3	1-3
Feldspar	3	0.5	1-3	2-5
Calcite / Siderite		5.6	0.6	0-2
Dolomite		1.3		
Analcite				
Pyrite	0.25	1.1	0.02	
Mica	4	1.4		
Illite	16	4.6		
Gypsum	0.7	0.4	0.14	
Rutile / Anatase	0.26	0.4		
Organic Matter	0.2		~0.3	
Other			0.8	

\*Includes 10-15% illite mixed with smectite.

As shown above, bentonites typically considered for use as buffer materials are montmorillonite-rich. Specifications for bentonites under development internationally (for example, reference [76]) consider the quantity of such swelling clay minerals, along with the type of charge compensating cations, which together control the swelling properties of the clay. Specifications also identify the maximum allowances for accessory minerals contained in the clay that can be detrimental to the long-term performance of disposal containers (primarily sulphide minerals, such as pyrite) together with specified physical properties such as the bentonite's dry density at a specified water resaturation level, hydraulic conductivity, and swelling pressure. The magnitude of the swelling pressure developed by a bentonite buffer will depend on factors that include:

- the composition of the bentonite
- the dry density of the bentonite at emplacement
- initial hydraulic conductivities and resaturation of engineered seals
- the composition of the host rock porewater.

A large number of experiments have been performed internationally to underpin understanding of how a bentonite buffer would resaturate [33, 77] and to measure bentonite swelling capacities (for example, see reference [78]). Bentonite considered for use in engineered barrier systems typically contains a high proportion of montmorillonite, a clay mineral belonging to the smectite family, which swells significantly when in contact with water. Figure 8 shows how different bentonites (that is, smectites containing particular counter ions) swell in deionised water [79]. In an unconfined system, smectite containing monovalent Na<sup>+</sup> or K<sup>+</sup> ions swells considerably, whereas smectites containing divalent ions (Ca<sup>2+</sup>, Mg<sup>2+</sup>) swell much less, although the difference is much smaller in a confined system such as an engineered barrier in a GDF. The amount of swelling is also controlled by the salinity of the groundwater, which controls the thickness of the double layer on the smectite grain surfaces and, thus, the osmotic pressure. A simplified atomic model of smectite is shown in Figure 7.

# Figure 7 Simplified atomic model of an idealised smectite structure [80].



Two parallel 2:1 layers are shown with vacant trans-octahedra (in reality, the stacking is irregular). Aluminium (light blue) is present in the octahedral sheet; magnesium and iron are also commonly found.

Swelling in dioctahedral clays is due to two types of swelling: innercrystalline swelling caused by the hydration of the exchangeable cations in the dry clay; and osmotic swelling, resulting from concentration gradients in ion concentrations between clay surfaces and pore water. Innercrystalline swelling is of prime importance under GDF conditions and

osmotic swelling is less significant. Osmotic swelling depends to a large extent on the electrolyte concentration and the valency of the dissolved ions, whereas innercrystalline swelling depends only slightly on these factors. Increasing the salinity of the pore water removes water from between the clay layers and decreases the swelling pressure.

Bentonite barriers will typically be constructed underground in a GDF using compressed blocks of partially resaturated bentonite. Some barrier construction schemes also involve the use of bentonite pellets or granules for filling small gaps between the host rock and the bentonite blocks. Determining the optimum geometrical proportions of bentonite with different characteristics (highly compacted blocks, pellets – see Box 5) that could be used to backfill GDF concepts that have large voids, such as concepts for multi-purpose containers, would require further work if these concepts are to be developed further. Internationally there has been a lot of work carried out to understand the impact of fabrication [81, 82], storage and the emplacement approaches (by pre-compacting the components, for example) for a chosen clay buffer/backfill material [83, 84]. Such investigations have also considered pre-saturation of bentonite once emplaced in order to rapidly fill the gaps in the emplacement tunnels and achieving the required performance with predictability.

As illustrated in Figure 8, water absorbed by bentonite that is able to physically expand causes swelling. If the bentonite is constrained and unable to expand freely, a swelling pressure develops, which locally reaches its peak at full water saturation. The swelling can be conceived of as being caused by a force of repulsion between the montmorillonite layers (see Figure 7).

# Figure 8 Variation in swelling capacity for a range of montmorillonite mineral samples.



Resaturation of the near field will cause bentonite buffer materials to become progressively more hydrated and this will cause the swelling clays they contain (specifically, smectite) to swell and, where confined, to develop increasing swelling pressures [30]. The hydration and swelling of these clays causes the barrier to develop mechanical properties that protect the waste packages from external influences such as earthquakes [29]. The barrier also develops low permeability that inhibits advective flow [30]. This limits the rate at which potentially corrosive groundwater species, such as chemically-reduced aqueous sulphur species, may migrate to the container surface, limits the activity and mobility of microbes, filters colloids, and limits the transport of corrosion products away from the container so that additional corrosion is not promoted [29]. Chemical reactions between the bentonite materials and the resaturating waters also condition the bentonite porewater environment, which promotes container longevity [33, 85, 86], see also subsection 3.9.1.

Resaturation of the GDF with chemically-reducing groundwaters would help to promote anaerobic conditions, thereby reducing the rates of corrosion and inhibiting many localised corrosion mechanisms. Reducing conditions will also lower the solubilities of many radioelements once released from the waste package [87]. As the thermal conductivity of saturated materials is typically higher than that of the dry materials this will also help to dissipate radiogenic heat in the near field.

Measured swelling pressures developed by MX-80 bentonite held at constant volume after water saturation are shown in Figure 9 [30]. Figure 9 (top) illustrates the influence of density and water salinity on the swelling pressures that develop. Once saturated, and depending on its initial density, the hydraulic conductivity of bentonite is very low, see

Figure 9 (bottom). It should be noted that the effect of salinity in reducing swelling pressure is only significant at low dry densities.

# Figure 9 Swelling pressure (top graph) and hydraulic conductivity (bottom graph) of MX-80 bentonite measured at different densities and molar concentrations of NaCl in the saturating solution [30].



Our understanding of bentonite resaturation has been further developed through studies modelling the Bentonite Rock Interaction Experiment (BRIE) at the Äspö Hard Rock Laboratory [88, 89]. The UK modelling team has been able to develop modelling approaches as part of a collaborative international project, providing us with the opportunity to:

 develop methodologies for calibrating models of fractured rock using site measurements as they become available • develop modelling techniques, allowing accurate representation of the interaction between the groundwater flow from the rock, and the resaturation of the bentonite material.

The approach has found that:

- the characteristics of bentonite resaturation are attributed to heterogeneous inflows, and accurate representation of the surrounding fractured bedrock is critical to understanding the hydration of emplaced bentonite
- the resaturation rate of bentonite in a fractured host rock is significantly affected by both the locations and total volume of groundwater ingress to the deposition holes investigated in the BRIE [88]
- the permeability of the rock matrix very strongly determines the prediction of the resaturation time of the emplaced bentonite, therefore this has implications for the characterisation requirements of the host rock [89]
- although models to date have been developed that are specific to the BRIE at the Äspö Hard Rock Laboratory, the tools, calibration techniques and methodologies developed are generic, and are directly applicable to any future site-specific investigations of bentonite hydration in a UK higher strength rock.

#### 3.7.2 Hydration and reaction of cement-based backfills

Cement-based materials will evolve through contact and reactions with groundwater; these materials contribute to the containment of waste by buffering porewaters to high pH.

Cement-based buffers and backfills will evolve through contact and reactions with groundwater. Through such processes, the cementitious materials will contribute to containment of the waste by buffering porewaters to high pH levels (described in subsection 3.10.1) that tend to slow corrosion rates and limit the solubilities of some radionuclides (as discussed previously). Elevated temperatures during the early post-closure period may cause some mineralogical changes in cementitious materials. Generally these changes include the development of more crystalline solid phases that are less soluble and might buffer to slightly lower pH values, but for longer periods of time. These changes can affect the pore structure and thus the hydraulic properties of the cement buffer/backfill. In addition, various mechanical effects (for example, cracking) may also occur [49].

#### 3.7.3 Dry conditions in an evaporite

An evaporite environment such as an undisturbed rock salt is extremely dry. Therefore, resaturation does not occur as it does in other types of host rock.

As a consequence of its low permeability and porosity, evaporite environments such as an undisturbed rock salt are extremely dry [3]. In general, salt formations would not endure in the neighbourhood of large amounts of flowing water. In regions of evaporite formations that might be identified as suitable for the GDF, the total amount of liquid present would be very low and dominated by brine internally trapped in the host rock, for example, as small fluid inclusions, brine pockets and intergranular moisture. Therefore, for concepts developed in an evaporite the term resaturation does not apply analogously to the process of resaturation may refer only to the filling of the crushed salt backfill pore space with brine before achieving complete compaction (discussed further in subsection 3.8).

Normal evolution for an engineered barrier system in an evaporite considers that the undisturbed host rock environment will remain effectively dry, with only local movements of brine inclusions or intergranular fluids under the influence of temperature gradients in the vicinity of heat-emitting waste packages. No radionuclide migration takes place through flow in connected transport pathways in this host rock.

Typically for such disposal concepts, the waste package is required to provide physical containment until creep closure is complete. In the early post closure phase dry near-field evolution is guaranteed by shaft and access-tunnel seals, hence, completeness of creep closure is not required. At a well-chosen site, appropriate performance of the shaft and access tunnel seals is essential for dry (normal) GDF evolution. Thus, proper functionality of these seals is the dominant factor in assuring acceptable performance after GDF closure. Considering a buffer/backfill that absorbs moisture and thus reduces the total amount of available moisture/brine is also an option, for example magnesium oxide in bags is stacked above waste containers at WIPP, see subsection 4.3.5. Compaction of crushed salt backfill is discussed further in subsection 3.8.

# 3.8 Compaction of crushed salt backfills

In evaporites, creep closure, or convergence, of the host rock is one of the key processes underpinning the long-term safety of a GDF. Over about a thousand years, the porosity of a crushed salt backfill in an evaporite GDF will be eliminated by compaction due to creep of the host rock and it will develop properties almost identical to those of the surrounding host rock. Elevated temperatures accelerate creep closure.

Evaporites may exhibit significant plastic flow, or creep. Creep will begin immediately following excavation in such rocks, driven by the presence of differential stresses due to the creation of void spaces. In evaporites, creep during the early post-closure period may cause the EDZ to evolve and recover. Over time, the porosity of the EDZ will be eliminated and the zone will develop properties almost identical to those of the surrounding host rock, merging with it to provide a continuous, effectively impermeable barrier [90].

Host rock creep is an important process to understand, particularly in evaporites, so that the components of the engineered barrier system that may be affected by it can be designed such that they provide the required functionality over a specified time period. The process of host rock creep is entirely site-specific and is discussed in more detail in the Geosphere status report [3].

In the illustrative concept for the disposal of HLW and spent fuel in an evaporite [91] (see also subsection 4.3), a crushed salt backfill is employed, which isolates the waste containers from porewater in the host rock. Host rock creep will lead to the compaction of crushed salt backfills and shaft seals used in disposal facilities constructed in evaporite host rocks [92]. Over time, the porosity of the crushed salt will gradually be eliminated – as the porosity reduces there will be a value at which the material will become impermeable and therefore meet its sealing requirement. Eventually the salt buffer and backfill components will develop properties almost identical to those of the surrounding host rock, merging with it to provide a continuous effectively impermeable barrier. Based on estimates for compaction of salt materials made for the illustrative concept for disposal of HLW and spent fuel in an evaporite, creep and compaction of tunnel and shaft seals will reach a permeability value similar to that of the host rock within approximately 200 years [91], the rate of creep being enhanced at elevated temperatures. Creep and compaction of the crushed salt buffer/backfill might take 1000 years or even longer. This will prevent water flow to the disposal region and prevent radionuclide release, thereby contributing to geological containment.

Bulk dissolution of salt in backfills used in disposal concepts designed for evaporite host rocks is not expected because there is no mechanism under undisturbed conditions to bring large volumes of low ionic strength waters into the tunnels. The brine inclusions and free intergranular brines that are sometimes present in evaporite rocks, could move locally around waste packages under the influence of a temperature gradient but would not concentrate into tunnels and are, in any case, already highly saline and therefore unable to dissolve further significant amounts of salt.

#### 3.9 Bentonite buffer (and backfill) evolution

For heat generating wastes, in many disposal concepts, bentonite clay is a key material used in the buffer and backfill. It may be emplaced as blocks of highly compacted bentonite or as pellets. Based on international laboratory and full scale demonstration experiments there is a mature understanding of the processes affecting bentonite buffer (and backfill) evolution, for example, dry density and the influence of intersecting fractures.

The bentonite buffer and backfill are key components of the engineered barrier system for heat-generating wastes in all geological environments except evaporites. The ability to manufacture and emplace the buffer and backfill to well-defined specifications, and the level of quality assurance required, is an area of active research in many European countries. Most emplacement systems are based on the use of prefabricated blocks of highly compacted bentonite, supplemented by the use of bentonite pellets or granules. Some experience of bentonite fabrication and emplacement is described in Box 5.

In clay-based concepts, once resaturation is complete, bentonite buffers and backfills will have reached their full swelling capacity; only regional gradients will persist and therefore hydraulic gradients will be much smaller than during resaturation. After this time, reactions occurring in the near field will affect the bentonite porewater chemistry, which in turn will promote or inhibit subsequent processes. In this way, evolution of bentonite porewater chemistry is intrinsically linked to the complex interactions occurring in the engineered barrier and has a strong influence on the rate of radionuclide release following container degradation in the far distant future.

Ideally, the bentonite porewater chemistry should have the following properties:

- low redox potential (Eh), providing reducing conditions
- relatively high pH, but not so high that it affects the stability of the bentonite [93]
- moderate salinity, to ensure that the bentonite remains stable and is not subject to
  excessive cation exchange (which might affect its swelling capacity) or chemical
  erosion (which can result from interactions with very dilute groundwaters)
- absence of aggressive species such as sulphides and chlorides or organic matter.

The subsections that follow explore how the properties of bentonite porewater are likely to evolve as a result of groundwater ingress and interactions with other engineered barriers. We then discuss several types of processes that might potentially affect the performance of bentonite barriers (and other engineered barriers) during the post-closure period. Such processes include:

- thermal alteration [79, 94, 95] (previously discussed in subsection 3.5.2)
- irradiation (previously discussed in subsection 3.6.1)
- erosion and piping [78, 94]
- ion-exchange [59]

- mechanical evolution [30]
- iron-bentonite interactions [96]
- hyperalkaline porewater-bentonite interactions [97, 98, 99, 100, 101, 102].

Each of these processes is considered in our forward programme (see Tasks 461-474 in our Science and Technology Plan [10]). The potential significance of such processes will depend on the site and disposal concept(s) chosen. Those processes listed above that have not already been discussed elsewhere are detailed in subsequent subsections.

# Box 5 Bentonite fabrication and emplacement

There have been many full-scale tests to emplace pre-fabricated bentonite blocks and pellets/granules, with both in situ underground trials and tests in tunnel mock-ups for both higher and lower-strength rocks. Rectangular blocks will typically be used for tunnel backfill, whereas shaped blocks will be used for the buffer. Pellets and granules will be used to fill gaps – for example, to manage irregular interfaces between blocks and the rock surface of a tunnel or disposal hole. For large caverns, in situ compaction of granules or powder might be used to emplace thick bentonite layers.

For the KBS-3V GDF concept for higher-strength rocks [103], the combination of different shapes of block and bentonite pellets deployed in the tunnels and deposition holes are shown below (Figure courtesy of SKB, Sweden).



On the left figure, 1 = backfilled tunnel, 2 = bentonite buffer, 3 = copper container, 4 = host rock.

The picture below shows shaped block emplacement being tested in both a mock-up opening (a) and underground (b: in granitic rocks in Switzerland) as part of the FEBEX project to simulate the axial, in tunnel container deposition concept of an engineered barrier system [104].



Handling and emplacement of annular buffer blocks for vertical container emplacement in higher-strength rocks are shown under test as part of an EC project (LUCOEX) [105] in the image below.



A common finding of all underground tests is that the emplacement of pre-compacted bentonite blocks is sensitive to local humidity in the environment and is not ideal for use in environments with high humidity. Under such conditions, the blocks can become friable and difficult to emplace without leaving voids or zones that do not meet the tight specifications required.

Consequently both humidity and localised water inflows need to be carefully managed during emplacement operations. Systems are being designed in Sweden and Finland to allow controlled water management in deposition holes during the buffer emplacement process. The current concept is to use a retractable rubber sleeve inside the deposition hole, within which bentonite blocks can be emplaced [106]. Such systems will need to be refined and tested for routine, industrial-scale application.

# 3.9.1 Mechanical evolution of bentonite

Mechanical properties of bentonite which contribute to its performance are a self-sealing ability, the swelling pressure and creep properties. It also needs to be sufficiently soft to offer protection to the container in the case of a rock shear arising from seismic activity.

The mechanical properties of bentonite are essential for its performance in many respects. The buffer backfill or seal will, most likely, be installed as a combination of blocks and pellets, but there will also be engineering voids in the system. Initially, the different components will have different properties.

To ensure that diffusion is the dominant transport mechanism in the buffer a low hydraulic conductivity is required. Since the installed buffer will have joints, a self-sealing

characteristic is necessary. A self-sealing ability is also required if fractures could be formed from extensive drying (or any other process).

In addition to the non-mechanical functions discussed elsewhere, the swelling pressure and creep properties are necessary to keep the container in place. There is an exponential dependence of the swelling pressure as a function of dry density of the material. The dimensions of the buffer need to be designed not to cause excessive mechanical loads on containers, liners, seals and the host rock.

The buffer also offers protection to the container in the event of rock shear resulting from seismic activity. Therefore, it needs to be sufficiently soft while the other favourable properties are maintained. In a repository in lower strength sedimentary rock, the host rock will interact mechanically with the buffer due to convergence. More details on some of these aspects are given in Section 3.4.1 of reference [30].

#### 3.9.2 Bentonite porewater

The composition of bentonite porewater can affect its evolution and will depend on the nature of the groundwater, but typical bentonite porewaters are rich in sodium, calcium and magnesium.

Bentonite porewater composition and evolution are strongly dependent upon the nature of groundwater entering the engineered barrier system. It will also be strongly dependent on the original composition of the clay. For example, reference [107, Table 7-2] shows that the content of soluble/partly soluble anions can vary rather substantially between bentonites of different geographical origin. Two conceptual models of solute transport through bentonite are discussed in Box 6.

Geochemical evolution of bentonite porewater is influenced by its interactions with montmorillonite surfaces (montmorillonite being the main constituent of bentonite), as well as interactions with other minor minerals present in bentonite through:

- dissolution and re-precipitation of montmorillonite and bentonite accessory minerals
- cation exchange reactions
- redox reactions and the solubility of resulting species
- container corrosion and the resulting corrosion products.

Such reactions will impact on the salinity of porewater, that is, the levels of different salts such as sodium chloride, magnesium and calcium sulphates, and bicarbonates, present in the porewater. Typical bentonite porewaters are rich in sodium, calcium and magnesium cations [104, 108], so salinity of the pore water changes as a function of the degree of saturation of the bentonite. As a consequence, saline fronts may be generated during hydration processes [109]. The mechanism of water uptake by bentonite is also affected by the salinity of the water. The resulting changes to the mineralogical composition of the bentonite can affect the desirable swelling properties of the buffer [109], although no significant alteration should occur before the bentonite is fully resaturated.

#### Box 6 Models of solute transport through bentonite

The transport of solutes through bentonite is important as it will determine the flux of any corrosive ions from the groundwater to the waste container and the migration of radionuclides from a perforated waste package through the buffer. There have been many measurements of mass transport through saturated, highly compacted bentonite. Generally the results show large dependencies on solute concentrations and bentonite densities, and striking differences in the transport capacity of anions and cations under certain conditions. Two different conceptual models exist to describe the observed behaviour: structural models, which include additional porosity with different properties to the basic montmorillonite interlayer pores; and a model based on Donnan equilibrium and the behaviour of charged species near a semi-permeable membrane.

In one conceptual model, compacted bentonite is viewed as possessing different types of porosity [110]. In this model, 'total porosity' refers to the total volume of voidage, without discrimination regarding location or type, whereas 'interlamellar/interlayer porosity' is located in the interlayer spaces of individual clay particles, between the individual tetrahedral-octahedral-tetrahedral sheets. This is considered to be a few monolayers thick and because of its more structured nature, is likely to have different properties from free water. 'External porosity' can be viewed as being of two types: that which consists of water in electrical double layers on the surfaces of the clay particles; and 'free water' or 'chloride porosity', which consists of water as interconnected thin films on the outside of clay stacks and also as films surrounding other minerals (for example quartz) in the bentonite. The amounts of each porosity type are thought to vary with compaction density in bentonite, with free/chloride porosity being significantly less than the total porosity as compaction density increases.

An alternative conceptual model [111] to that described above consists of a bentonite-pore fluid system consisting of one main porosity type and where pore fluid composition is controlled by ion equilibria within the interlamellar pore space involving two basic processes: Donnan equilibrium which reduces concentrations of external ions compared with external pore fluids; and cation exchange, which affects systems only with more than one type of cation. Consequently, the clay-pore fluid system is envisaged to consist of clay particles acting as macro-ions, and the entire clay-water system may be viewed as a 'polyelectrolyte'.

In general, increasing salinity of groundwaters that are taken up by bentonite as it resaturates tends to reduce the swelling pressure, which is an adverse effect with respect to the buffer's desired functionality. However, this effect is less apparent at increasing initial density of the bentonite and it can be concluded that, for the target densities envisaged for typical buffer applications, groundwater salinity has only limited significance. This can be observed in experimental results such as those shown in the top graph in Figure 9, which shows the swelling pressure vs dry density of MX-80 exposed to different concentrations of sodium chloride solution.

Montmorillonite, one of the main mineral constituents of bentonite (see subsection 3.7.1, Box 4) has a high cation exchange capacity which, in particular, enables it to sorb caesium and strontium [112]. When contacted with groundwater entering the near field, cation exchange reactions can take place between the cations in montmorillonite and ions present in the groundwater. Such reactions are more likely to occur between bentonite and high salinity groundwaters.

The information relating to the composition of the groundwaters at GDF depth obtained during site investigations will be important for understanding the importance of bentonite alteration processes, such as the transition of smectite to illite. Transformation from montmorillonite to illite is well documented in several different geological environments and

has been reproduced under laboratory conditions [30]. The mechanisms underpinning this alteration are complex, but at the temperatures expected in the GDF, the alteration of smectite to illite is likely to be slow [30].

#### 3.9.3 Redox potential of bentonite

Reducing conditions in a bentonite buffer will be established relatively quickly through the consumption of oxygen by corrosion, reaction with minerals in the bentonite buffer and host rock, through microbial action and due to the ingress of reducing groundwaters. Low redox potential is important in order to minimise the rate of spent fuel container corrosion.

Redox conditions in the engineered barrier system will initially be oxidising as a result of the presence of air trapped in the engineered barrier system at closure. Following closure, reducing conditions in a bentonite buffer will be established relatively quickly through the consumption of oxygen by corrosion, reaction with minerals in the bentonite buffer and host rock, through microbial action and due to the ingress of reducing groundwaters [72, 95, 113, 114, 115].

Further materials present in the near field may take part in redox buffering reactions (such as trace sulphide minerals present in bentonite and iron/steel components of the engineered barrier system); reactions with these materials also tend to buffer low porewater redox potentials. Microbially-mediated reactions may influence redox conditions and, again, these reactions will tend to drive the system towards increasingly reducing conditions. In more detail, the redox potential of porewaters in the engineered barrier system may be buffered at low (reducing) levels by one or more redox couples (for example, Fe(II)/Fe(III) and U(IV)/U(VI)). Not all redox reactions will attain equilibrium and, spatially, there may be a range of redox conditions within the GDF. However, studies of redox potential in deep disposal facilities suggest that conditions will generally be reducing [116, 117, 118]. Work undertaken by SKB estimated the time taken for oxygen in the near field of a KBS-3V disposal facility (that employs a bentonite buffer) to be consumed at between 10 and 300 years [51, 119]. Investigations of the buffer material from the prototype repository [120] indicate that reducing conditions prevail in the buffer in even shorter timeframes. Similarly, redox evolution modelling performed by Nagra estimated that reducing conditions would develop within 100 years of closure [33], owing to corrosion of steels and the presence of minerals such as iron sulphide (pyrite) present in their Opalinus Clay host rock.

It should be noted that redox evolution in the buffer will differ according to whether copper or steel waste packages are employed. Reduced sulphur species are the main corrodants of copper and need to be minimised. In other words, the presence of pyrite in bentonite may be good for creating reducing conditions, but this benefit is offset against the generation of copper corrodants. Steel is a much more chemically-reactive material than copper and will impact significantly upon redox evolution, gas generation, and mineralogical alteration of the buffer. Copper will be much more benign.

The development of reducing conditions within the GDF tends to slow further container corrosion and causes redox-sensitive radionuclides to be present in their lower redox states, which in turn tend to be less soluble and, therefore, less mobile.

Under some circumstances relatively more oxidising glacial meltwaters might reach the GDF [29], but this is only potentially relevant to the late post-closure period because glaciation of the UK is not thought likely until significantly more than a hundred thousand years (with significant uncertainty) in the future [3, 8]. Also, radiolysis reactions in the engineered barrier system may produce small amounts of oxidised species [30], which could have subsequent effects on the chemistry in the buffer (for example, see Box 7).

# Box 7 Impacts of radiolysis on porewater redox potential

Radiolysis of water in the near field can generate oxidised species and hydrogen gas. Such species could potentially influence redox conditions in the near field. The spatial distribution of redox potential in the engineered barrier system will be dependent on the reducing capacity of materials present within each of the materials and on the specific location within the disposal area. For example, containers with iron content will corrode and provide corrosion products capable of controlling redox potential (via the Fe<sup>2+</sup>/Fe<sup>3+</sup> redox couple); bentonites have a far lower reducing capacity, since only traces of reactive iron minerals are present (for example, siderite and iron oxides).

The effect of released radiolytic oxidants on redox conditions in bentonite buffers has been calculated for a range of disposal concepts. For systems employing steel based waste containers for spent fuel, coupled mass balance models ([121] and references therein) have indicated that, under pessimistic assumptions  $H_2$  is non-reactive and passivation of magnetite formed from the corrosion of iron-container materials limits the release of iron into solution. For such a system, the container environment could become oxidising, which can influence the rate of fuel dissolution (see Behaviour of Radionuclides and Non radiological Species in Groundwater status report [4]), but the largest part of the bentonite would remain reducing.

Impacts of radiolysis have also been considered with respect to bentonite buffers and their porewaters in systems considering more corrosion-resistant container materials [30]. In such systems, the production of water radiolysis products, including  $H_2O_2$ ,  $O_2$  and  $H_2$  are estimated to have low significance for the long-term stability of bentonite [30]. Such evaluations suggest that radiolysis of bentonite pore water will be insignificant as the dose rate outside the container will be too low to have any effect [30]. Similar conclusions were reached for the effects of radiolysis in the backfill (as the radiation field is even lower in these regions).

# 3.9.4 pH buffering capacity of clay materials

In general, bentonite tends to have a buffering effect on the near-field pH to neutral or mildly alkaline values, but does not buffer the pH to the highly alkaline values promoted by cementitious materials.

The pH evolution of bentonite porewater may be determined through the interaction of a number of factors, such as: ion exchange on clay; protonation-deprotonation reactions at clay edge sites; dissolution-precipitation reactions of trace carbonate minerals (calcite, siderite, dolomite); dissolution-precipitation reactions of the major clay mineral component (montmorillonite) of the bentonite; the concentration of anions (usually chloride) in the groundwater; and the partial pressure of carbon dioxide of the system. The consensus is that the clay fraction principally acts as a cation exchanger, with the clay silicate exchanger being essentially inert and pH being determined by the contribution from the trace carbonate mineral concentration, and the ambient chloride activity. In natural systems smectite clays may undergo dissolution-precipitation reactions over assessment-relevant timescales at pH 9-10 and temperatures of 50-60 °C [122]. Thus, clay hydrolysis reactions make a significant contribution to pH buffering in bentonite over the long-term.

In general, bentonite may be considered to have a buffering effect on the near field pH to neutral or mildly alkaline values (for example, pH values between pH 7.2 to 9.4 [123]). This is controlled mainly by carbonates in the bentonite, but also occurs due to dissolution and re-precipitation of other bentonite accessory minerals and by protonation and deprotonation reactions at the surface of bentonite minerals [109]. Although the carbonate content of

bentonite tends to be relatively low, the associated dissolution and precipitation reactions are crucial to its ability to buffer the alkalinity of bentonite pore waters [116].

Bentonite does not buffer the pH to the highly alkaline values generated by cementitious materials such as NRVB, which minimise the solubility of many elements. Nevertheless, this behaviour has a minor beneficial effect on the evolution of the near field and on the performance of the engineered barriers. For example, pH values between 7.2 and 9.4 are such that copper corrosion rates are low and are not increased as they might be by the inflow of more acidic (for example, sulphide-bearing) lower-pH groundwaters (see the Waste Package Evolution status report [2]). In this pH range smectite and other clays in the bentonite are not subject to alteration as they might be at more alkaline (higher) pH values (see subsection 3.9.7).

#### 3.9.5 Piping and erosion of bentonite buffers

If there is high water inflow to the GDF local to a bentonite barrier the water pressure may act on the emplaced bentonite and piping erosion may occur; this can lead to the formation of channels in the bentonite. Bentonite might also be eroded in dilute waters as a result of chemical processes that lead to bentonite swelling and the formation and dispersion of colloids.

Resaturation is unlikely to occur evenly across the entire GDF. This may particularly be the case if flowing fractures or other localised water-bearing features contact individual disposal holes for HLW and spent fuel disposal containers. If fluid flow through such features is pronounced, then some near-field barriers (such as those composed of bentonite) may be susceptible to piping erosion [124] or chemical erosion processes.

Piping erosion (a mechanical process) can occur if flow in the region of the buffer-rock interface is high, caused by elevated hydraulic gradients in the early period after emplacement; chemical erosion can occur if dilute waters come into contact with bentonite – for example, as a result of changing environmental conditions affecting deep groundwater chemistry (see Box 7). Figure 10 below illustrates a photograph of an experiment performed using bentonite to investigate piping processes [125]. The photograph shows a channel formed by piping during a test in which a 1% salt solution was passed at a flow rate of 0.1 l/min through bentonite pellets in a Plexiglas tube [125].

In order to ensure that bentonite buffers perform their function, it is necessary to take account of the range of host rock conditions that may be encountered in the GDF during barrier installation. At some locations in the GDF the excavations may be relatively dry, while at others, particularly in fractured rocks, there may be significant water inflows that can cause piping erosion of emplaced bentonite materials. These processes have the potential to remove significant amounts of bentonite from a deposition hole and hence could reduce the ability of the buffer to protect the container from chemical and mechanical degradation processes [79]. Under extreme conditions of erosion, advective flow of groundwater might develop in the buffer region. Piping and erosion processes are discussed further in Box 8.



### Figure 10 Photograph of piping of bentonite.

Photograph of a channel formed by piping during a test in which a 1% salt solution was passed at a flow rate of 0.1 l/min through bentonite pellets in a Plexiglas tube.

To reduce the possibility of such processes affecting the performance of a bentonite buffer, each individual excavated disposal hole will be assessed and monitored for water-bearing rock fracture zones and mechanically-weak rock prior to waste emplacement. Typically this comprises application of rock characterisation methods based on geophysics and geohydrology. Such approaches are currently being examined by SKB in addition to other engineering solutions [79, 30, 126] and methodologies are being developed by Posiva to define criteria for the screening and acceptance of disposal hole locations [127]. Similar research would be required to consider the effect of site-specific conditions in the UK on piping and erosion if a fractured higher strength host rock were identified as host rock for the GDF.

The EC BELBaR project, looking at the impacts of bentonite colloids [128], is further investigating bentonite erosion. The BELBaR project aims to understand the main mechanisms of erosion of clay particles from the bentonite surface and to quantify the (maximum) extent of possible erosion under different physico-chemical conditions. These studies have investigated under what conditions compacted bentonite is able to produce colloidal particles, free to move into the contacting aqueous phase. Data obtained at a laboratory scale are being compared to those obtained in-situ at the FEBEX gallery at the Grimsel Test Site where a real-scale experiment, simulating an HLW repository in granite, was installed over a decade ago. The BELBaR project also aims to validate and advance the conceptual and mathematical models used to predict mass loss of clay in dilute waters and clay-colloid generation, as well as clay-colloid facilitated radionuclide transport relevant to geological disposal of higher level radioactive waste [128]. The BELBaR project is discussed further in the Behaviour of Radionuclides and Non radiological Species in Groundwater status report [4].

#### Box 8 Piping and erosion processes

#### Piping

Bentonite piping is the formation of hydraulically conductive channels in bentonite-based engineered barriers due to groundwater flow under local hydraulic pressure gradients around disposal holes and tunnels. Bentonite materials are likely to be emplaced in the GDF as pre-compacted blocks and pellets. The bentonite materials emplaced will only be partially saturated with water (the extent depending on the fabrication and compaction technique). After emplacement, the bentonite will take up more water from the surrounding rocks and will swell.

If water inflow to the GDF is localised in fractures that carry more water than the swelling bentonite can adsorb, the water pressure will act on the emplaced bentonite. Where the gel is too soft to stop water inflow, piping erosion will occur and can lead to the formation of channels in the bentonite (as shown in Figure 10). The channels formed during piping have a strong effect on focusing water flow and this focussed flow may facilitate further erosion of the bentonite. Bentonite piping is influenced by water flow rates, the properties of the bentonite or bentonite-based materials used, and by water salinities and compositions. Piping is considered to be a feature applicable to the early period after waste emplacement, before disposal tunnels are backfilled and sealed; SKB consider that piping will not occur after a GDF is fully resaturated because hydraulic gradients will have reduced [29].

Nevertheless, some piping could affect a large number of container deposition locations during the operational period, before disposal tunnels are backfilled and sealed, and Posiva has estimated [129] that roughly one third of container positions in the Olkiluoto GDF design could be affected to some extent. Posiva observes that the exact physical process of piping remains unknown and the theoretical understanding is being further developed.

#### Chemical erosion in dilute groundwaters

Bentonite might be eroded as a result of chemical processes that lead to bentonite swelling and the formation and dispersion of colloids at the bentonite-water interface, or if water flows are fast enough to cause shearing at the bentonite water interface, leading to the formation of colloids or larger particles.

Over the last few years, large research programmes in Scandinavia have investigated bentonite erosion [79] under conditions of varying water salinity and flow rates. If bentonite is contacted by dilute water, salts initially in the bentonite porewater may diffuse out into the surrounding water and be dispersed. The ion concentration at the bentonite-water interface may then decrease and a stable colloidal suspension of clay can form due to the repulsive forces between the clay particles [79].

The formation of stable suspensions of bentonite colloids could lead to clay being eroded from a bentonite barrier in a GDF if groundwater flow rates are high enough and groundwater salinities are low for long periods of time. This situation might occur, for example, if there were to be a prolonged period of temperate climate with progressive invasion of dilute waters to depth. Erosion is more likely to occur in fractured host rocks with active, dilute groundwater flow systems than in systems with lower groundwater flow (such as in un-fractured host rock environments) or with more saline waters [79]. As an example, at the Olkiluoto GDF site, Posiva [129] estimate that a period of greater than 200,000 years of dilute water penetration would result in more than 10% of container locations experiencing dilute conditions. This kind of estimation is highly dependent on many factors (see below), including the nature and properties of the fracture network in which flow occurs and the measures taken to define acceptable container locations.

For either piping or chemical erosion to affect performance of the engineered barriers, large

amounts of buffer material (of the order of hundreds of kilograms in a single container deposition location) would need to be eroded before significant advective flow of groundwater could occur. In some disposal concepts this might lead to rapid waste container corrosion.

The likelihood of occurrence and the significance of bentonite erosion will depend on:

- the disposal concept
- the design of the engineered barrier system
- the specific bentonite materials used
- the pattern and rates of groundwater flows within the host rock and the EDZ
- the chemistry of groundwaters and how these might evolve with time at GDF depths
- the chemical interaction between groundwaters in fractures and immobile porewaters in the rock.

#### 3.9.6 Iron interactions with bentonite

Iron-bentonite interactions could occur in some disposal concepts. Two mechanisms may be envisaged: the alteration of montmorillonite to iron-rich smectite and the replacement of smectite with non-swelling phyllosilicates. Most assessments assume the physical properties and transport properties of bentonite remain unchanged despite these interactions.

There are a number of possible disposal concept examples where iron-bentonite interactions could occur; primarily when using bentonite buffers together with iron-based components (such as steel construction materials and steel waste-container materials). Under post-closure GDF conditions, steel present in engineered barriers will corrode slowly to produce a range of possible steel corrosion products [2]. Steel corrosion products such as magnetite, iron carbonates, iron sulphides and iron (oxy)hydroxides will be formed under chemically-reducing conditions, dependent on the dissolved carbonate, chloride and sulphide concentrations in the vicinity of the steel surface [130]. Rather than forming a thick layer of corrosion products (as thought previously), these corrosion products can then react with bentonite by substituting smectite interlayer cations with dissolved iron(II) cations [131, 132, 133].

Two potential bentonite alteration mechanisms may be envisaged [134]:

- the alteration of montmorillonite to iron-rich smectite
- the replacement of smectite with non-swelling phyllosilicates, such as chamosite (iron-rich chlorite) or berthierine/odinite (1:1 iron-rich minerals).

There may also be redox effects in the original montmorillonite by the reduction of Fe(III) to Fe(II), which could lead to a change in the layer charge, eventually affecting the stability of the mineral.

Increased concentrations of iron in bentonite will therefore affect some of the physical properties of bentonite, such as increased hydraulic conductivity and possibly decreased swelling pressure.

The sorption capacity of the bentonite could become saturated with an excess of iron (either as Fe<sup>2+</sup> or FeCl<sup>+</sup> in chloride-rich solutions) since iron sorption is strong on clay edges [135], such that sorption sites in bentonite could be blocked by iron prior to container penetration and release of radionuclides. This could have an impact on the transport of released radionuclides through the bentonite following wasteform leaching or dissolution.

Most assessments assume that the physical properties (swelling and self-healing) and the transport properties (diffusion and sorption) of bentonite remain unchanged despite ironbentonite interactions. Work continues to develop further understanding of iron-bentonite interactions so that the longevity of the buffer to maintain the necessary swelling capacity is achieved. Work is ongoing within our research programme to develop understanding of the implications of this process on near-field system evolution for a range of generic concepts (see Tasks 462 and 463 in our Science & Technology Plan [10]). This understanding, in isolation from any specific disposal concept, will support material optioneering for concept development activities.

#### 3.9.7 Hyperalkaline porewater interactions with bentonite

Reaction of hyperalkaline porewaters (originating from cementitious materials in the GDF) with bentonite could alter the bentonite mineralogy through processes such as cation exchange, smectite dissolution and precipitation of calcium carbonate. Such reactions may result in changes in bentonite porosity, swelling pressure, hydraulic conductivity and sorption capacity.

There are several interfaces in an engineered barrier system where hyperalkaline porewater (cement conditioned waters at pH > 12) can react with bentonite (and other clay-based minerals), resulting in changes in the bentonite properties [136]. The most important interfaces to consider include reactions due to:

- hyperalkaline porewaters originating from cementitious materials such as fracture grouting, shotcrete, and tunnel plugs used for either mechanical stabilisation of tunnels, or to seal fluid transport features [137]
- hyperalkaline porewaters originating from a cement-backfilled ILW/LLW disposal area [137].

Reaction of hyperalkaline porewaters with bentonite can alter the bentonite mineralogy through processes such as cation exchange, smectite dissolution and precipitation of calcium carbonate. Such reactions may result in changes in bentonite porosity, swelling pressure, hydraulic conductivity, and sorption capacity, and hence its ability to fulfil a containment function, as described in Box 9.

# Box 9 Hyperalkaline porewater reactions with bentonite

The most important processes in defining the spatial and temporal extent of hyperalkaline porewater-clay interactions and consequent changes in bentonite properties that could impact on the buffer functions are [100, 138, 139]:

- diffusive transport of cement-pore fluids into bentonite, with mixing and reaction with the clay-pore fluids. Sharp gradients in pH (and pCO<sub>2</sub>) across the interface encourage the rapid precipitation of solid carbonates, such as aragonite and calcite, and hydroxides such as brucite (Mg(OH)<sub>2</sub>), leading to a decrease in porosity, although this may not have a significant impact on bentonite functionality.
- fast exchange of cations in cement pore fluids (principally K<sup>+</sup>, Na<sup>+</sup>, and Ca<sup>2+</sup>) for cations (mainly Na<sup>+</sup> in bentonite) in interlayer sites in montmorillonite, leading to a decrease of swelling pressure.
- slow dissolution of montmorillonite and other minerals present, such as quartz, feldspars, pyrite, and gypsum. At elevated pH, such reactions consume hydroxyl ions, thus chemically neutralising the advancing cement pore fluids. These reactions lead to an increase in porosity and may decrease clay swelling pressure due to mass loss.

Hyperalkaline porewater interactions with bentonite have been studied extensively in the last ten years through laboratory experiments, natural analogues and modelling [140]. However, most of these studies have considered the interaction of OPC-type cement conditioned porewaters with bentonite (and not formulations of low-pH cements).

Where bentonite is present as a barrier or backfill material, concerns about its potential interactions with cement conditioned porewaters have led most, if not all, waste management agencies<sup>11</sup> to adopt 'low-pH cements' (see Box 10) for all uses of cementitious materials (for example, fracture grouting, shotcrete, tunnel plugs) in the vicinity of a bentonite buffer or barrier [79, 141, 142]. Moreover, most agencies have active programmes to limit and account for all uses of cement and concrete in any disposal environment where cementitious leachates may contact bentonite [143]. For example, in higher-strength rocks, experiments are being carried out in Sweden and Finland to use diamond-wire rock sawing or reaming to prepare smooth disposal-tunnel floors, rather than employing a concrete tunnel base. This has the added benefit of potentially reducing the EDZ in the tunnel floor.

# 3.10 Cement backfill evolution

Cement backfills are typically employed to condition incoming groundwater to a high pH (typically pH > 9) and to have an extensive buffering capacity that maintains these conditions for an extended period of time.

Box 10 considers some uses of cementitious materials in underground engineering. Cement backfills used in engineered barrier systems provide an important barrier to radionuclide migration.

<sup>&</sup>lt;sup>11</sup> For example, Andra in France, SKB in Sweden, Posiva in Finland, Nagra in Switzerland and JAEA in Japan.

#### Box 10 Use of cementitious materials in underground engineering

Cementitious materials are widely used in underground engineering. In a GDF, specific consideration has to be given to:

- the interaction of hyperalkaline porewaters with vitrified waste (affecting its solubility see the Waste Package Evolution status report [2]),
- deterioration of the properties of bentonite buffers and tunnel backfills, and
- chemical reactions with alumino-silicate minerals in the host rock (see the Geosphere status report [3]).

The deleterious effects of leachates from (for example) Ordinary Portland Cement (OPC) are dominantly caused by sodium, calcium and potassium hydroxides released from the cement into the leachate, which is consequently hyperalkaline, with a pH >12 and up to 13.5. In order to minimise these interactions, 'low-pH' cement formulations are considered, where leachate pH values below about 11 are targeted. The application of low-pH cement formulations has also been demonstrated in full scale tests as part of the EC ESDRED project. During the optioneering process for materials selection, consideration would also be given to the feasibility of cement emplacement at an industrial scale using the shotcreting technique for rock-wall lining. The photographs below show shotcreting of an excavation tunnel (a) and low pH shotcrete panels (b).



Photographs showing application of shotcreting of an excavation tunnel (a), and preparation of low-pH shotcrete panels (b) 1396-01-Esdred/Seidler/NDA Through a combination of chemical retardation processes (solubility limitation, precipitation, co-precipitation and sorption, see Box 11), a cement backfill contributes to the objective of containment. The combination of these processes, promoted by a cement-based backfill chemically conditioning the environment of the near field (in this context), is sometimes referred to as 'chemical containment' or a 'chemical barrier'.

### Box 11 Chemical containment

The equilibration of cement minerals with groundwater will establish alkaline conditions through the dissolution of alkali metal hydroxides and, in particular, calcium hydroxide from the cement. These alkaline conditions form part of the basis of chemical containment [146].

Chemical containment of radionuclides in cement-based barriers is achieved by a combination of [4]:

- **solubility limitation**, which acts to provide an upper limit to the dissolved concentration of the radionuclide in the water in contact with the wasteform. This is the maximum possible concentration that may be attained. For some radionuclides, the solubility limit is sensitive to the water chemistry; in particular the pH and redox conditions, and the absence or presence of organic complexants.
- precipitation, co-precipitation and sorption, which act to remove radionuclides from solution and hence reduce their migration through the engineered barrier system.

Cements developed for use as backfills in geological disposal are designed against specified requirements. These requirements are derived from the performance needed from a cement backfill in a specific disposal concept. For example, the Nirex Reference Vault Backfill (NRVB), as described in Box 12, was developed for application in a higher strength host rock. NRVB was specified to fulfil a number of requirements (such as facilitating package retrievability using high-pressure water-jetting), including providing conditions for chemical containment [52]:

- long-term maintenance of alkaline porewater chemistry to suppress dissolved concentrations of many important radionuclides, under the prevailing hydrogeological conditions of the disposal area
- long-term maintenance of a high active-surface area for the sorption of key radionuclides
- relatively high permeability (relative to the host rock) and porosity to ensure both homogeneous chemical conditions (so that localised concentrations of materials in wastes will not exhaust the desired chemical conditioning and thereby locally reduce the containment performance) and to permit the escape of gas generated by chemical reactions within the disposal area.

Similarly, other waste management organisations have developed and characterised high porosity cement backfills, designed to provide high porosity for gas storage and transport while still withstanding high axial stresses [144]. In contrast, cement materials for disposal area tunnel linings and/or for shotcreting are developed with enhanced requirements for their mechanical properties - such as low gas accessible porosity and low permeability (typically  $1 \times 10^{-17}$  m<sup>2</sup> [144]). As outlined in Box 10, waste management organisations are also developing low alkaline cements for potential use in tunnel linings, shotcretes and backfill.

#### Box 12 Nirex Reference Vault Backfill

The Nirex Reference Vault Backfill (NRVB) was developed for the illustrative UK ILW/LLW disposal concept in a higher-strength host rock.

The NRVB was designed to provide the alkaline conditions and sorption capacity that are the principal features of chemical conditioning in the UK's ILW/LLW GDF concept, and thereby the provision of a chemical barrier to radionuclide migration from the engineered barrier system. To this end, the pH buffering performance of the backfill is required to persist for time-scales normally considered in performance assessments - that is, for a period of time up to a million years following its emplacement in the GDF. In practice, this requirement should be met if the backfill provides pH buffering via the dissolution of calcium hydroxide and calcium silicate hydrate gel from a material based on Portland cement.

In addition to the chemical and mineralogical requirements, the NRVB has been designed to be emplaced in suitable volumes with minimal bleed (excess standing water produced during hydration). In addition, the material has a high porosity to facilitate sorption, the maintenance of uniform chemical conditions throughout a GDF and the release of gas generated from the waste [52, 145].

The Nirex Reference Vault Backfill has been designed to fulfil these requirements. The backfill comprises ordinary Portland cement with a fine aggregate containing crushed limestone filler and hydrated lime (calcium hydroxide). The formulation is:

Component	Weight fraction	
Ordinary Portland Cement	0.26	
Fine Limestone Aggregate	0.29	
Hydrated Lime Aggregate	0.10	
Water	0.35	

The backfill has a porosity of about 0.5 and a saturated density of 1730 kg m<sup>-3</sup>. The high porosity means that the compressive strength of the material is low; less than 6 MPa after 28 days of curing, facilitating retrievability via high-pressure water-jetting (prior to closure). A more detailed discussion of the requirements used during the selection and design of NRVB can be found in references [52, 145, 146].

The evolution of cement-based backfills in the engineered barrier system of a GDF disposal area will be driven by a number of processes that may occur under post-closure conditions [147]. These are discussed in the following subsections and may be summarised as:

- dissolution of cements in groundwater and reactions with groundwater solutes and the host rock, locally affecting rock properties (by, for example, pore clogging)
- the transformation of metastable minerals to more stable and crystalline forms, including continuing hydration of cements
- the possible development of significant spatial heterogeneity in the disposal area through, for example, cracking in the backfill, precipitation of new minerals and the development of preferential groundwater flow
- possible reactions between the cementitious backfill and other engineered barriers
- reactions between cement backfills and wastes or, more significantly, the products of waste degradation.



# Figure 11 Evolution of a cement-based engineered barrier system.

These processes, and the resulting evolution of the physical and chemical properties of a cement backfill, will depend on the prevailing conditions (for example, temperature and pressure), the extent of groundwater flow into the engineered barrier system, any creep or other mechanical deformation or rock collapse caused by stresses in the host rock, and various chemical processes, including chemical reaction, mineral alteration, precipitation

and dissolution. Figure 11 illustrates how we expect a cement-based engineered barrier system to evolve over post-closure timeframes.

A near-field component model is currently under development for a cementitious concept for disposal of ILW and LLW. A component model is a model that includes a number of different processes in a subsystem (in this case the engineered barrier system), and may feed information to the total system model used to calculate system performance in the post-closure safety case. The near-field component model couples a reactive transport model of the evolution of the cement backfill with a probabilistic representation of radionuclide transport through the near field and will allow for a more realistic treatment than is possible in the performance assessment models. More detail on the future development of the near-field component model can be found in our Science and Technology Plan, tasks 444 and 446 [10].

The following subsections discuss various aspects of the evolution of cement backfill. A recent review of the use of cement materials in geological disposal concepts [56] provides more detail on some of these subjects; the topics covered by the review include:

- the emplacement of cement materials as backfill
- the chemistry, types and properties of cementitious materials
- the long-term mechanical performance of cements
- the effect of temperature on the performance of cements
- the effect of groundwater on the leaching and pH-buffering of cement backfills
- the effects of interactions with other engineered barrier system components on cement backfills
- the sorption properties of cementitious materials
- the development of novel low pH cements for structural concretes and sealing applications.

#### 3.10.1 Cement leaching and pH buffering

When cement is mixed with water various hydration products form, the most common including calcium-silicate-hydrate gels of variable composition and Portlandite. The leaching of cementitious materials by groundwater at long timescales can reduce the pH of the cement porewater.

When cement is mixed with water various hydration products form. Depending on the chemical composition of the cement and the water, and on whether any other materials (for example, gypsum or calcite) are present, the most common hydration products include calcium-silicate-hydrate (CSH) gels of variable composition and Portlandite (calcium hydroxide, Ca(OH)<sub>2</sub>), plus a range of other less abundant solid phases such as ettringite, monosulfate, monocarbonate, and hydrotalcite [148].

The solid phases in a hydrating cement are typically in contact with (and may incorporate) porewater. Long-term chemical reactions between such porewaters and the hydration products result in the partial dissolution of the solid phases present. As an example the reaction between Portlandite and water can be written as follows:

 $Ca(OH)_{2(s)} + H_2O_{(l)} = Ca_{(aq)} + 2OH_{(aq)}$ 

It can be seen that dissolution of Portlandite produces hydroxyl ions (OH<sup>-</sup>), which cause the water to become highly alkaline. Experimental studies show that this reaction is exothermic

and reaches equilibrium quickly. At room temperature this yields water containing ~20 mM  $Ca^{2+}$  with a pH of ~12.5.

The other cement hydration products also dissolve in water to produce alkaline solutions. The dissolution of CSH gels yields waters with pH values in the range ~10 to 12.5, depending on their composition and the prevailing conditions.

In situations where there is a sufficient mass of cementitious materials containing such hydration products these types of reaction will dominate and control, or buffer, the chemistry of the porewaters present. As waters flow into and through the cementitious materials more of the solid phases dissolve and the chemistry of the porewaters is, thus, held constant, or buffered by the appropriate composition. Figure 12 illustrates a simplified description of the sequential stages of dissolution of cement in pure water and expected pH buffering.

The dissolution of cements in groundwater (as opposed to de-mineralised water) has also been modelled using chemical modelling software, based on models of the chemistry of calcium silicate hydrate (CSH gels) in pure water [147, 149].

Natural analogues of hyperalkaline conditions and rock-water reactions similar to those in, and caused by, OPC leachates have been studied over many years [150]. A location at Maqarin in Jordan in particular has yielded much information on the nature and timescales of the reactions shown in Figure 12 and their impacts on the surrounding rock [151].

# Figure 12 The evolution of pH at 25°C in cement pore fluid as a result of cement degradation in pure water. Taken from [152].



As part of our research programme numerous modelling studies have been undertaken to develop our understanding of the dissolution of cements, including NRVB [147, 153].

Leaching of NRVB follows the same sequential stages of cement dissolution as illustrated in Figure 12, with each later stage potentially lasting many thousands of years, depending on the hydrogeological environment:

- Stage I: Leaching of highly soluble alkaline hydroxides to give high pH porewater.
- Stage II: Dissolution of Portlandite (calcium hydroxide) to buffer the porewater to approximately pH 12.5 at 25°C.

- **Stage III**: Incongruent dissolution of a solid, meaning that the ratio of the concentration of ions in solution is different from that in the solid. In the case of incongruent dissolution of calcium silicate hydrate (CSH) gel, the calcium is leached preferentially from the solid and the Ca/Si ratio of the solid is gradually reduced until a congruent point is reached. The congruent point being when the Ca/Si ratio in the solid is equal to that in solution. Congruent dissolution of CSH phase then occurs until it is depleted.
- **Stage IV**: Dissolution of the lower solubility minerals such as calcium carbonate.

An overview of the properties and performance of cementitious materials in a GDF is given in reference [52]. The validity of this description has been demonstrated by the results of leaching experiments for cementitious materials [154].

Depending on groundwater composition, reaction of groundwater solutes with dissolved cement minerals will likely be accompanied by the precipitation of new minerals [22]. For example:

- carbonation will occur, that is, carbonate in groundwater entering the near-field through resaturation, or formed through the dissolution of carbon dioxide gas arising from the microbial degradation of organic wastes [5], can react with calcium hydroxide to form calcium carbonate (CaCO<sub>3</sub>) (in addition to any limestone already present in the cement backfill [52])
- magnesium in groundwater can react with calcium-rich minerals to form magnesium hydroxide (brucite, Mg(OH)<sub>2</sub>); this compound may also be produced from the corrosion of Magnox cladding waste
- aluminium may react with calcium and sulphate in groundwater to form sulphate minerals such as ettringite [155, 156].

Such reactions may influence both the evolution of the mineralogy of the solid and the composition of the cement-equilibrated groundwater. The potential significance of such reactions will depend on the composition of the groundwater, the concentrations of the solutes, and volume changes associated with changes in the mineralogy. The spatial distribution of such precipitates may also influence the evolution of heterogeneity of a cement-based engineered barrier system.

The formation of precipitates may change the performance of the cement by forming protective layers on the backfill surfaces and modifying the pH conditioning capacity. For example, leaching and/or carbonation of cementitious materials will lead to a gradual lowering of porewater pH from ~12.5 down to values of ~10, depending on groundwater chemistry and rates of groundwater flow. The effect of these processes may be to increase or decrease backfill and EDZ porosity through fracture armouring, described in Box 13. Such effects may alter the way in which the backfill resaturates and may alter permeability locally [46].

A number of research projects aiming to improve our understanding of aspects of the carbonation of cementitious materials have recently been completed. This process is of interest because of the resulting changes to the physical and chemical properties of the materials and also because of the ability to remove carbon-14 containing carbon dioxide from the gas phase. One study has investigated the reaction kinetics of carbonation under a range of conditions; the rate of carbon dioxide uptake was shown to be dependent on the nature of the material and the degree of water saturation, as well as the partial pressure<sup>12</sup> of carbon dioxide [157]. A further study has investigated the carbonation of 20-year old

<sup>&</sup>lt;sup>12</sup> In a mixture of gases, each gas has a partial pressure which is the hypothetical pressure of that gas if it alone occupied the volume of the mixture at the same temperature.

blended cement pastes [158], proposing a process model of carbonation occurring in hardened cement pastes.

A further experimental programme has assessed the carbonation behaviour of NRVB [159]. Carbonation capacities calculated from small scale trials were in agreement with previous studies. Large scale gas experimental trials showed that the NRVB does retard the progress of any released carbon dioxide by carbonation [159].

# Box 13 Crack healing through carbonation of backfill

The precipitation of new minerals, for example calcium carbonate, due to reactions between the cementitious materials and groundwater solutes is likely to occur in small cracks (typically ~2 to 5 mm in size [46]) that make up the fracture network within a cement backfill. In such circumstances, the crack may be partially or completely closed, a process commonly called 'crack healing' [160].

Crack healing has been observed in the Maqarin natural analogue study, with ettringite formation occurring in fractures in the rock affected by the natural hyperalkaline plume (see picture below – ettringite is denoted 'Ett': source: A. E. Milodowski, British Geological Survey).



Crack healing behaviour has been found to be strongly influenced by the nature of the cementitious materials involved, as well as depending on the concentrations of the groundwater solutes. The current state of knowledge is however insufficient to demonstrate that all cracks within the near field would be healed permanently.

The dissolution of cements with groundwater to produce high-pH porewaters contributes to chemical containment and also supports containment provided by the waste package through iron passivation, thereby slowing the corrosion of iron and steel waste containers [161].

To underpin our understanding of cement dissolution over the long-term post-closure timeframe, many research studies have been performed in the UK and overseas to investigate the details and specifics of cement hydration, and these studies have led to the

development of sophisticated chemical models of cement porewater chemistry [148, 162, 163, 164, 165, 166].

An illustration of recent modelling work undertaken as part of our research programme is shown in Figure 13 [167]. Here the TOUGHREACT program has been applied to predict the spatial and temporal changes in the chemical conditions of an ILW/LLW disposal area, see reference [22]. This is an example of a coupled reactive transport model that is capable of taking into consideration the cement used as an encapsulation grout in typical UK ILW waste packages, together with a candidate cement backfill material (NRVB), and consideration of the interaction of these cement materials with a range of typical groundwater compositions (a saline groundwater example is shown in Figure 13).

Reactive transport models such as that illustrated in Figure 13 have been used to make estimates of the length of time cementitious materials may buffer the chemistry of waters in the GDF. In general, studies such as these suggest that under expected near-field conditions cementitious backfills will buffer the chemistry of porewaters to high pH values for at least thousands, and probably for hundreds of thousands, of years [46, 147, 154, 164, 167, 168, 169].

Models for the leaching of cementitious materials by groundwater are used to estimate the rate of change of chemical conditions. Sufficient quantities of backfill can be specified to take into account the effects of groundwater leaching, such that GDF porewater remains sufficiently alkaline to provide a 'chemical barrier' for a long time. The quantities of backfill to be used in the GDF are based on a consideration of reactions that consume calcium hydroxide and which apply a number of cautious assumptions about the reactions between different cements and between cements and wastes (or their degradation products) [170]. A more realistic treatment would probably result in a lower requirement for the amount of NRVB in the GDF. Consideration of such methods is currently being developed and applied to model the dissolution of candidate cement backfills (see Tasks 428 and 429 in our Science and Technology Plan [10]).

Figure 13 Predicted spatial evolution of pH over a period of 50,000 years (for groundwater with a 'Saline' composition flowing through NRVB backfill surrounding a 3:1 Blast Furnace Slag (BFS)/Ordinary Portland Cement (OPC) grout.



#### 3.10.2 Cracking of the backfill

A source of heterogeneity in a cement-based engineered barrier system will be the inevitable cracking of the cement backfill. Cracking of the backfill may facilitate a more rapid flow rate through a disposal vault, but this may be balanced by the potential for cracks to heal by chemical interactions and the precipitation of mineral phases.

An important source of heterogeneity in a cement-based engineered barrier system will arise from cracking of the cement backfill. The presence of a network of cracks may affect the transport of radionuclides out of the GDF and the chemical conditioning of the backfill porewater (as discussed in subsection 3.10.1).

As described in subsection 3.10.1, together with the feasibility of emplacement of a cement backfill in a disposal area, consideration will also be given to minimising any degradation of the properties of the cement backfill prior to closure. However, some backfill shrinkage and cracking are likely to be unavoidable. These processes will continue during the early post-closure period prior to the complete resaturation of the backfill.

Cracking of cements starts shortly after emplacement, predominantly as a result of their exothermic hydration reaction and also as a result of mechanical stresses resulting from the thermal expansion of the engineered barrier system materials, the host rock and near-field porewater [49]. The processes that potentially cause cracking are discussed further in referenced [46].

Experimental evidence suggests that the presence of a network of cracks can dominate the mass transport behaviour of a cementitious material [147]. Any cracks that are formed in a cement backfill will tend to channel groundwater flow. Therefore, in a cracked environment it is unlikely that all of the disposal area backfill volume will be accessed by flowing groundwater, and the transport of material between cracks may be dominated by a diffusion process that is much slower than the advective rate of water flow in the cracks.

The conditioning of the chemistry of groundwater flowing in a cementitious ILW/LLW disposal area where the cementitious materials have cracked has been the subject of scoping studies using simple models [171]. These studies suggest that it is possible for the pH of flowing water in cracks in cement to be lower than expected for the porewater in uncracked cement

A summary of the results of previous studies investigating the cracking of NRVB is provided in reference [52]. In the current, generic, phase of our programme we have been building on this work, focussing on understanding better the spatial heterogeneity in the near field of a cement-based backfill. Such studies that have been reported recently include developments in the understanding of cement cracking mechanisms (for example, cracking from early age plastic settlement and thermal contraction, and the expansion of waste packages as a result of corrosion) [46]. This work has modelled the development of preferential groundwater flow and its chemical evolution through a representative network of cracks in the backfill, including estimates for the likely concentrations of dissolved species in, and the pH of, groundwater as it flows through cracked backfill. Conclusions from this most recent work (noting that uncertainties are associated with predicted values for all modelling activities) indicate that significant cracking of a cementitious backfill will occur:

 during emplacement of the backfill, due to its plastic settlement under solid horizontal waste package surfaces; this could be expected to result in horizontal gaps (cracks) of up to 2 mm under the base of each package. These cracks would extend only as far as the edge of the waste packages, so will not be connected. Gaps are less likely to occur under stillages, which have holes in the base plate that allow the flow of backfill and bleed water.

- within days of backfilling, through early-age thermal contraction of the backfill, which will result primarily in vertical cracks of up to 0.2 mm. These are likely to extend between waste packages at locations where the backfill is thinnest. Some of these cracks may connect the gaps previously formed under waste packages.
- post-closure, due to the expansion of waste packages (and associated gas generation), which could result in horizontal cracks of up to 5 mm; cracks of up to 0.1 mm may occur before closure. These could extend across the full width of the vault between each layer of waste packages, expanding the gaps under packages generated due to settlement of the backfill and connecting adjacent gaps.
- potentially, as a result of vault roof collapse if the crown space is unfilled, or if the vault contents progressively compact, or due to the impact of earthquakes over the long post-closure period.

Cracking of the backfill from such processes may facilitate a more rapid flow rate through the centre of a disposal vault [52], but assessment of this effect must be balanced by consideration of the potential for cracks to heal by chemical interactions and the precipitation of mineral phases, as discussed above (see Box 13).

#### 3.10.3 Redox evolution in cement systems

Most groundwater that will flow into the facility will be reducing. Its redox potential will continue to be influenced by steel corrosion products, maintaining reducing conditions in spite of the oxidising effects of any radiolysis.

Various processes will influence the redox potential of porewater in a cement backfill. In general, these processes will tend to consume any oxygen initially present in air within the disposal area, or in porewaters, and will cause chemically-reducing conditions to develop [72, 114, 115]. Such processes include corrosion, particularly of ferrous metals (see Box 14), and the inflow of reducing groundwaters. The depth envisaged for geological disposal facilities in the UK is such that most groundwaters that will flow into the facility are likely to be reducing [172]. A possible exception to this is that under some circumstances relatively more oxidising glacial meltwaters might reach the GDF, but this is only potentially relevant to the late post-closure period because glaciation of the UK is not thought likely for at least the next hundred thousand years or so [3].

The redox potential will be largely influenced by the corrosion of steel and the presence of steel corrosion products, which are assumed to maintain reducing conditions once they are established. The redox potential will influence the solubility and sorption of a number of radioelements and chemotoxic metallic species. Typically, the solubility of most of these redox-sensitive elements tends to be lower, and the sorption stronger, under reducing conditions (lower redox potential) where lower oxidation states of the elements are thermodynamically more stable [4], examples being radioisotopes of technetium and neptunium [121]. References [173, 174, 175] provide details on the current understanding concerning the inter-relationship between corrosion and Eh evolution, and the consequences for the solubility and sorption of redox sensitive radioelements.
### Box 14 Fe(II) / Fe(III) redox buffer

The reducing capacity of most ILW/LLW disposal systems is very large, particularly because of the large quantity of steel (in the form of structural supports and waste containers). Steel corrosion will lead to the formation of magnetite, which is the thermodynamically stable iron phase under expected near-field conditions.

We are able to estimate redox evolution in a disposal system with large quantities of steel by considering iron(III) and iron(II) equilibria. The three main Eh controlling processes are described below. These occur in the sequential order of:

- 1. Eh controlled by the generation of Fe<sup>2+</sup> by steel corrosion and the precipitation of goethite (FeO(OH)).
- 2. Eh controlled by the transformation of goethite to magnetite ( $Fe_3O_4$ ).
- 3. Eh controlled by the precipitation of magnetite. The Eh in this period will tend to the Eh of the equilibrium Fe(0)/magnetite. During this period the H<sub>2</sub> generated is accumulated in solution until it reaches the pressure threshold of the system. Once achieved, H<sub>2</sub> gas escapes from the system and the Eh becomes constant with time.

Steel corrosion is likely to be the principal process governing redox conditions. Other redox couples may also contribute to reducing conditions being maintained (for example U(IV)/U(VI)), however not all redox reactions will attain equilibrium and, spatially, there may be a range of redox conditions within the GDF.

Other near field materials may take part in redox buffering reactions (for example, blast furnace slag in cementitious grouts and trace sulphide minerals present in bentonite) and reactions with these materials also tend to buffer low porewater redox potentials. Microbially-mediated reactions may also influence redox conditions and, again, these reactions will tend to drive the system towards increasingly reducing conditions [22].

Modelling studies of redox potential carried out for geological disposal facilities suggest that conditions will generally be reducing [114, 173]. Modelling work has also taken into consideration the impacts of radiolysis of water [33, 72]; conclusions from this work reported that hydrogen gas generation expected as a result of radiolysis will be insignificant when compared to that from the microbial degradation of organic materials and the corrosion of mild steel containers and metallic wastes.

Widespread anaerobic conditions are expected to be established shortly after closure of the disposal area as a consequence of the aerobic corrosion of steel [173]. If it is assumed that equilibrium thermodynamics apply in the longer term, reducing conditions will be maintained at a calculated redox potential (Eh) relative to the standard hydrogen electrode (SHE) of about -450 mV (at pH 12.5 and 25°C) by the iron(II)/iron(III) couple [176]. In addition to the principal redox governing process of steel corrosion, as outlined above, recent modelling work [173] has considered a range of additional near-field system features and coupled processes that could potentially impact the overall redox potential in the wastes and in the cementitious backfill. Such coupled processes include how the rate of metal corrosion and the variations in groundwater composition can influence the evolution of the oxidation potential. Conclusions from this work have indicated that neither a regional groundwater conditioned by a clay environment nor more oxidising groundwaters would significantly change the expected Eh evolution. The same conclusions were drawn with regard to different steel corrosion products, their reactivity and the surface area of steel available for corrosion. However, the results reported show that the corrosion rate greatly influences the timescale of the different redox buffer periods identified, and also the time at which the final redox potential is achieved, but not the Eh value of the different periods.

Microbial activity may also influence the extent to which conditions are reducing, at least in locations where microbial populations become established. Active microbial populations can mediate the reduction of some oxyanions (see subsection 3.13), and there is evidence for their reduction of dissolved metal ions (such as iron(III) and uranium(VI) [177]). The influence of microbes on redox potential will be considered in our ongoing research programme, as shown in the Science and Technology Plan [10], tasks 392, 443, 773, 766 and 768.

### 3.10.4 Reaction of cement backfills with other cementitious materials

The evolution of the chemical barrier in a cement-backfilled disposal area is primarily based on our knowledge of pH conditioning through the dissolution of the backfill. However, the waste encapsulation grouts used in packaging of some wastes are also cementitious materials and may also contribute to the pH conditioning.

Our understanding of the evolution of the chemical barrier in a cement-backfilled disposal area is primarily based on our knowledge of pH conditioning during backfill dissolution [178, 179]. However, the waste encapsulation grouts used in packaging of some UK ILW are also cementitious materials, and may contribute to the conditioning of the pH of the near-field pore-water, or provide sites for radionuclide sorption.

Typical waste encapsulation grouts used in the UK (for example those based on a 3:1 mixture of pulverised fuel ash (PFA) and ordinary Portland cement (OPC), or on a 3:1 mixture of blast furnace slag (BFS) and OPC) are substantially different from cement backfills (like NRVB). They contain less calcium and more silicon, aluminium and iron, giving rise to a different mineralogy that commonly lacks calcium hydroxide and contains more calcium aluminate hydrate minerals [168]. Such mineralogy provides alkaline conditions, although the initial pH will be lower than that provided by NRVB and the evolution of its pH will differ [168].

The differing compositions of NRVB and the encapsulation grouts may provide a driving force for a slow reaction between the materials, ultimately resulting in a more uniform mineralogy. This reaction has been termed a 'pozzolanic' reaction, by analogy with the so-called pozzolanic reaction between dissolved calcium hydroxide and amorphous silica to form CSH gel, as observed in many cement systems [164], see Box 15. Furthermore, the porosity of encapsulation grouts in pristine conditions will tend to be lower than that of the NRVB, potentially hindering both the achievement of equilibrium solution chemistry and radionuclide sorption [44].

### Box 15 Pozzolanic reaction

The term pozzolanic is taken here to refer to inorganic materials that are capable of reacting with calcium hydroxide, or with materials which release calcium hydroxide, to form a product which hardens in water. Pozzolans react with calcium hydroxide in a hydrating cement system, forming CSH gel. The chemistry of the reaction products is very similar to those formed by typical OPC-based cement hydration, with small differences in the amounts of the resultant cement phases. Generally the Ca/Si ratio of the CSH gel is lower than that formed by hydration of OPC-based cement. Complete reaction of the calcium hydroxide may occur eventually, however the rate of reaction tends to fall as a layer of hydration products accumulates around the pozzolanic particles. It is essential that the pozzolanic material is in the form of small grains for reaction to occur.

PFA, silica fume and zeolitic material are all be capable of reacting with calcium hydroxide in the near field.

Further details regarding the pozzolanic reaction and how this has been previously assessed with respect to UK grouts and NRVB are available elsewhere [52]. In general, in determining the backfill requirement, the reaction is considered to remove calcium hydroxide from NRVB, so extra NRVB has been added to the GDF design to compensate for this loss.

Further details on the various waste encapsulation grouts typically used in the UK are provided in the Waste Package Evolution status report [2].

## 3.10.5 Reactions between cement-based engineered barriers and the natural barrier

At the interface between cement backfill and the host rock, dissolution of cementitious materials is likely to be accompanied by the precipitation of new minerals from the reaction of groundwater solutes with the dissolved cement minerals.

At the interface between cement backfill (and other cement-based engineered barriers) and the host rock, dissolution of cementitious materials by groundwaters is likely to be accompanied by the precipitation of new minerals that result from the reaction of groundwater solutes with the dissolved cement minerals. Where this occurs in grouted fractures and engineered tunnel linings, the resultant secondary phases may seal the fracture and effectively block flow locally. The potential significance of such reactions will depend on the composition of the groundwater (a site-specific factor), the concentrations of cement-derived solutes, flow rates, and volume changes associated with mineralogical changes.

Such a process could lead to an effective sealing (known as 'armouring') of the outer boundary of the cement backfill, and other cement material where in contact with the host rock. This would lead to decreased porosity and permeability locally in the excavation disturbed zone [180] which would impact on processes such as resaturation since groundwater flow (and subsequent cement degradation) would be slowed [33] due to a decreased diffusion rate. The excavation disturbed zone is discussed further in the Geosphere status report [3].

Evidence for the potential self-sealing of a cement-based ILW/LLW GDF is well documented [181]. Figure 14, taken from [181], illustrates the various porewater transport pathways through a cementitious near-field, indicating alternative transport pathways given effective sealing of the host-rock-near-field interface (Route A). The dotted lines indicate an initial background flow field that might be changed due to mineral reactions and porosity and hydraulic conductivity changes.

### Figure 14 Indications for self-sealing of a cement-based ILW/LLW GDF.

A: Sealing of the cement so that groundwater flow would be diverted around the cavern.

B: Porosity and hydraulic conductivity increase allows for additional groundwater flow through the cavern. C: Initial flow field.



### 3.10.6 Reactions between the cement and wastes

Reactions of wasteform grouts and wastes with a cementitious backfill can produce changes in the mineralogy and the associated performance of the backfill / grout. For example, organic wastes containing cellulose are susceptible to alkaline degradation, which produces soluble compounds which may form complexes with disposed radionuclides and inactive species.

The chemical environment of a cement-based engineered barrier system will have a significant impact on materials present within the waste (such as UK ILW). Organic wastes containing cellulose, such as paper, wood and cotton, are susceptible to alkaline degradation [182]. This is important because alkaline cellulose degradation produces soluble organic compounds able to form complexes with some radionuclides, enhancing their migration [183]. Alkaline degradation of cellulose generates a range of organic compounds with erythro and threo isomers of isosaccharinic acid (ISA) being the most abundant. Box 16 provides details on the key cellulose degradation products formed via the alkaline degradation of cellulose and the key near-field controls on this process.

### Box 16 Generation of cellulose degradation products by alkaline hydrolysis

The cellulose contained in ILW is likely to experience alkaline, anaerobic conditions [184] at temperatures of 30 to 60°C [26]. Under these conditions, degradation occurs predominantly by a mechanism called the 'peeling reaction', in which the cellulose degrades from one end of the chain of glucose units [22]. The peeling reaction results in the production of a range of water-soluble cellulose degradation products [185, 186]. Key amongst the degradation products, because of their complexing abilities and yields, are the (a) erythro, and (b) threo isomers of 2-C-(hydroxymethyl)-3-deoxy-D-pentonic (isosaccharinic) acid (ISA). Both isomers are illustrated below:



### Isomers of isosaccharinic acid (ISA)

The concentrations of cellulose degradation products in the waste will be reduced by chemical, radiolytic and/or microbial degradation, as well as by processes such as dilution or sorption once they have moved out of the waste package. Controls on the extent of degradation to produce ISA include temperature and the presence of oxidising agents within the wasteform.

The generation of cellulose degradation products and their impact on radionuclide mobility are discussed further in the Behaviour of Radionuclides and Non radiological Species in Groundwater status report [4]).

A considerable amount of work has been carried out to investigate the rate of the alkaline degradation of cellulose in the context of a cementitious GDF both in the UK (see, for example, reference [182]) and abroad [183, 187]. The combined effects of  $\alpha$ -radiolytic alkaline degradation of cellulose on plutonium leaching and solubility (relevant to plutonium-contaminated material (PCM) wastes) [188] and the effects of  $\gamma$ -irradiation and alkaline degradation have also been investigated [189]. A recent review [190] summarises our current knowledge concerning the rate and extent of cellulose degradation with time under typical GDF conditions and the identification of degradation products and the stability of those species (in particular ISA) under near-field and geosphere conditions.

In addition to degradation of organic material in waste packages to give carbon-based acidic products such as carbon dioxide and low molecular weight organic acids [191], other important reactions of wasteform grouts and wastes with a cementitious backfill to produce a change in mineralogy or a change in performance properties (for example, sorption properties) include:

- radiolytic degradation of polyvinyl chloride (PVC), yielding hydrochloric acid.
- some possible organic degradation products can interact with cements to form relatively insoluble precipitates, for example calcium carbonate or organic salts such as oxalates [22]. The formation of soluble calcium salts would accelerate the rate of

dissolution of the cements. For example, calcium aluminate hydrate minerals are dissolved by hydroxy-carboxylic acids [191].

- interaction of cements with non-aqueous phase liquids (NAPLs) such as oils and solvents that are present in existing wastes, or with NAPLs that may be generated by the degradation of some solid organic materials, such as plastics [192].
- carbonation of the wasteform. Reaction of wasteforms with dissolved carbon dioxide formed from the degradation of organic materials in the waste. This reaction reduces the alkalinity of the cement [164, 170].
- Siliceous materials (for example ion exchangers such as clinoptilolite) that may participate in a pozzolanic reaction.
- Aluminium and aluminium corrosion products that may contribute to the formation of calcium aluminate hydrate minerals and that may react with sulphate in the groundwater.

These potential reactions can be addressed in the design of a cement-based disposal area by the use of an excess of cement backfill over that required to buffer the pH in the absence of such acidic species [52].

In addition to cement-based wasteforms, other materials are considered for ILW and LLW encapsulation. We are doing work to develop our understanding of vitrified ILW and polymeric wasteforms and their expected evolution, see the Waste Package Evolution status report [2] and tasks 601, 602 and 586 of our Science & Technology Plan [10].

We have also carried out work to determine the fate of non-aqueous phase liquids (NAPLs) in ILW and LLW wasteforms. RWM has now concluded, on the basis of multiple lines of evidence, that significant quantities of NAPLs will not escape from waste packages, and that any NAPLs that do escape are unlikely to accumulate in such a manner that could result in them being transported into the geosphere [193].

### 3.11 Seals and plugs

Temporary and permanent seals and plugs will be required in areas such as individual deposition holes, at the end of cavern-type excavations, and for closure of access ways to waste disposal areas, such as the access tunnels, shafts and drifts. They aim to reduce groundwater flow or reduce potential pathways for groundwater flow.

The importance of seals and plugs in drifts and shafts is realised for most geological disposal concepts. This is an active area of international research, development and demonstration in which RWM is participating. Depending on the exact disposal concept, a range of temporary and permanent seals and plugs will be required in waste disposal areas and at the end of cavern-type excavations (ILW/LLW disposal concepts), in addition to use for closure of access ways to waste disposal areas, such as the access tunnels, shafts and drifts.

During construction and operations, some degree of sealing may be required at the end of excavated regions of the host rock (whether this be a disposal tunnel or cavern-style excavation) to provide temporary isolation (for example, using shotcrete) or a more permanent long-term degree of isolation (for example, using bentonite and/or concrete plugs). Such seals may need to be placed in strategic positions with respect to water inflow, as well as practical design aspects, such as the need to create separate (isolated) disposal areas. Figure 15 shows a drift sealing concept of a large volume low pH concrete plug from the French disposal concept [194].

For bentonite-containing disposal concepts, seals and plugs may be utilised to avoid problems when emplacing the buffer and the disposal containers, particularly where water inflow might create non-uniform swelling of bentonite. The installation of tunnel plugs will also prevent piping of bentonite by reducing hydraulic flux. Figure 16 shows the conceptual design for a deposition tunnel plug from SKB [195]. Figure 17 shows casting of a low pH concrete plug for the containment wall of the FSS (Full-Scale Seal) Experiment (see Box 17).

Complete sealing systems may also be required in order to close disposal areas once the wastes and the engineered barriers have been emplaced. Typically, for most concepts this involves high integrity sealing of the access tunnels and the access shafts to provide complete isolation of the GDF. For example, in evaporite rock concepts, sealing systems in the drifts and shafts are an important part of the safety concept to ensure that potential pathways for groundwater ingress are removed.

### Figure 15 Drift sealing concept of a large volume low pH concrete plug (shotcrete and cast concrete) for use in the French disposal concept



### Figure 16 Conceptual design for a deposition tunnel plug from SKB



## Figure 17 Casting of low pH concrete plug for the containment wall of the FSS Experiment



Materials used for seals and plugs will be dependent on the host rock environment and the degree of containment required from such materials over specified timeframes.

We recently participated in the Euratom's Seventh Framework Programme Full-Scale Demonstration of Plugs and Seals (DOPAS) Project. This focused on the demonstration of tunnel, drift, vault and shaft plugs and seals for crystalline, clay and salt rocks. The main experiments are described in Box 17 [196]; since completion of the EC supported phase some of these experiments will continue to yield data for some time to come. The project also consisted of a large number of complementary modelling and laboratory testing tasks which have been used to support instrumentation of the experiments or to confirm in situ conditions.

### Box 17 DOPAS Plug and Sealing Full-Scale Demonstration Tests

The EC DOPAS project focused on the demonstration of tunnel, drift, vault and shaft plugs and seals for crystalline, clay and salt rocks. The project consisted of five work streams:

**DOMPLU (DOMe PLUg Experiment) – Deposition tunnel plug:** Based on SKB's KBS-3V reference design, and installed at the Äspö Hard Rock Laboratory in Sweden, the DOMPLU plug consists of a dome-shaped, unreinforced concrete plug, a watertight seal and a filter zone. The function of the concrete plug is to resist deformation and to keep the watertight seal, filter and backfill in place. The watertight seal is made of bentonite blocks and pellets. Its function is to seal water-leakage paths and to ensure an even pressure on the concrete. The filter is made of sand or gravel. Its function is to collect water draining from the deposition tunnel so that no water pressure is applied on the concrete plug before it has cured and gained full strength. The DOMPLU experiment will help to reduce uncertainties in the long-term performance of deposition tunnel plugs. Specific objectives for the experiment included further development of water tightness requirements on deposition tunnel plugs and gaining plug production experience.

**POPLU – Deposition tunnel plug:** POPLU is testing an alternative tunnel plug design for use in the KBS-3V system at the ONKALO underground rock characterisation facility in Olkiluoto, Finland. The plug consists of a wedge-shaped reinforced concrete structure containing grouting tubes and circular bentonite strips at the rock-concrete interface to ensure water tightness. In addition, a backfill layer is planned behind the concrete structure to enable the pressure-testing of the plug. The majority of the requirements on this design focus on how the deposition tunnel plug contributes to post-closure safety - by keeping the backfill in place during the operational phase and ensuring that the plug does not significantly affect the post-closure performance of the backfill.

**EPSP (Experimental Pressure and Sealing Plug) – Deposition tunnel plug:** EPSP is an experiment on a tunnel plug at the Josef URL in the Czech Republic. Unlike DOMPLU and POPLU, the focus of EPSP is on gaining fundamental understanding of materials and technology, rather than testing of a reference or alternative design. EPSP consists of a pressure chamber, an inner concrete plug (the primary sealing component), a bentonite zone, a filter and an outer concrete plug. The primary sealing component is the inner concrete plug. Key aspects of the experiment are to evaluate the use of fibre reinforced sprayed concrete for the concrete plugs and sprayed bentonite pellets composed of Czech bentonite for the bentonite zone.

**FSS (Full-Scale Seal) – Drift and ILW disposal vault seal:** FSS is a full-scale (above ground) experiment of the reference drift and disposal vault seal for the French Cigéo repository concept. The seal consists of a cast concrete containment wall, a swelling clay core and a shotcrete containment plug. The experiment is focused on the technical feasibility of constructing a full-scale seal that is designed to limit groundwater flow between the underground installation and overlying formations, and to limit groundwater flow speed within the disposal area.

**ELSA – Shaft seal:** ELSA is a programme of laboratory tests that was used to further develop the reference shaft seal for the German reference disposal concepts for repositories in salt and clay host rocks. The design for the shaft seal, which is developed for the site-specific conditions at Gorleben, included three short-term sealing elements designed to maintain their functionality until the backfill in the repository drifts, access ways and emplacement fields has sealed in response to compaction driven by host rock creep. These sealing elements are a seal located at the top of the salt rock and made of bentonite, a second seal made of salt concrete, and a third seal made of sorel concrete (a magnesium oxide/chloride non-hydraulic concrete) located above the disposal level.

### 3.12 Gas generation

Key gas generation processes in the GDF will be the corrosion of metals and microbial action. Once generated, gas may dissolve in water, undergo chemical reactions or form a separate gas phase which may or may not be able to migrate out of the near-field.

Following closure of the facility, gas generation<sup>13</sup> will continue throughout the post-closure period [5]. Typically, non-radioactive gases that could be formed in large volumes (bulk gases) will arise from:

- corrosion of metals (in the wastes, the containers and the structural components of the GDF) in the absence of oxygen, giving rise to hydrogen production
- microbial degradation of some organic materials, yielding mainly methane and carbon dioxide
- radiolysis of water and organic materials, yielding mainly hydrogen.

The potential role of microbial activity is discussed further in the following section, but hydrogen is expected to be the main gas formed, with lesser amounts of methane and carbon dioxide. In addition, small amounts of helium will be generated as a result of the decay of alpha-emitting radioisotopes, and radioactive gases, including tritiated hydrogen and methane, inactive methane and carbon dioxide containing carbon-14, radon and krypton-85 will be released [5]. The volume of hydrogen generated will be very much greater than that of the radioactive gases. The quantities and rates of gas generation for UK wastes are described in more detail in the Gas status report [5].

Once generated, gas that is formed may dissolve in water, undergo chemical reactions or form a separate gas phase [5]. The effect of gas generation will depend on its ability to migrate through the near field and enter the geosphere. Gas will also interact with the near-field barriers as it migrates from the point of generation. The ease with which it can migrate will depend on the properties of the materials present (porosity, permeability), the degree of saturation and the properties of the gas (solubility and chemical reactivity). Potential impacts of gas on the evolution of the near field include:

- carbon dioxide generated from the microbial degradation of organic wastes can react with cement materials in the waste package or backfill (see subsection 3.10.1)
- hydrogen gas generated from radiolysis or corrosion processes may impact redox conditions in the near field (see subsection 3.9.3)
- micro-fissuring of bentonite due to an increased gas pressure (see Box 18)
- disposal tunnel plugs and seals, designed against gas transport requirements (such as those being considered in the illustrative concept for disposal of ILW/LLW in a lower strength sedimentary host rock [144]) will impact on the spatial and temporal evolution of a disposal area, and particularly the way in which a disposal area may resaturate.

The quantity of gas that is generated, its effect on the engineered barrier system, and its migration through the engineered barrier system will depend strongly on the disposal

<sup>&</sup>lt;sup>13</sup> Gas will also be generated during the operational phase of the GDF. Carbon dioxide produced from microbial degradation of (for example, cellulosic) waste materials is assumed to react with the wasteform grouts. Any unreacted CO<sub>2(g)</sub>, together with hydrogen (predominantly) gas produced from corrosion (for example, waste packages), will be removed by the ventilation system during the operational phase.

concept and the host rock. In some circumstances there may be no separate gas phase; in others, a separate gas phase may form and may be able to migrate out of the near-field easily (for example, in fractured higher-strength rocks), in yet other cases a separate gas phase might form and could become trapped in the near-field until the formation pressure is exceeded and a pathway for gas migration can open [5].

The impacts of gas generation on the evolution of the engineered barriers are taken into consideration throughout our engineered barrier system research. As our programme moves forward and concept development progresses more detailed understanding of the coupled processes, particularly between how gas generated in the engineered barrier system can influence the chemical and physical buffer and backfill evolution, will be developed.

Gas generation and migration were the focus of the integrated, multidisciplinary, EC FORGE project, with collaborative research between international radioactive waste management organisations, regulators and academia, specifically designed to tackle the key issues associated with the generation and movement of repository gases [197]. FORGE addressed these issues through a series of laboratory-scale and field-scale experiments, including the development of new methods for upscaling allowing the optimisation of concepts through detailed scenario analysis; FORGE is discussed further in the Gas status report [5].

### Box 18 Micro-fissuring of bentonite

Once generated, the ease with which gas can migrate through a bentonite buffer will be determined by the degree of buffer resaturation. Experimental observations suggest that a gas phase can migrate through an initially water-saturated buffer clay only if the gas pressure exceeds a threshold value (or gas-entry pressure, which has been related to the sum of the clay's 'swelling pressure' and the water pressure) [198]. It has been suggested that at some threshold gas pressure, the gas will create micro-fissures through the bentonite, a process termed dilatancy-controlled gas flow or pathway dilation [144].

However, confined bentonite at high saturations has a large swelling pressure and a commensurately large gas entry pressure. The gas entry pressure is so large that two phase flow, accompanied by the displacement of water, cannot occur. The only transport mechanism available in this case is diffusion. If the gas pressure exceeds the pressure in the bentonite then consolidation of the bentonite (the bentonite is compressed) and/or the formation of dilatant pathways occurs. The extent of consolidation will be limited and at some critical pressure pathways can form and gas will then migrate through these pressure induced micro fissures [5].

### 3.13 Microbial effects

Conditions in the GDF may be favourable for certain species of microbes. Some have the potential to alter the near-field chemistry and to promote corrosion and other degradation processes. Others may seal fractures or consume bulk gas. There is uncertainty surrounding the implications of microbial activity in the engineered barrier system.

Conditions in the GDF may be favourable for certain species of microbes to thrive [62]. Some species may be present naturally in the host rock [62]. Others may be introduced from the surface during GDF construction and operation. Many of these species will be inactive, or will have no significant effect on engineered barrier system evolution. However some, such as sulphate-reducing bacteria, have the potential to alter near-field chemistry and can promote corrosion and other degradation processes given the appropriate circumstances [199, 200]. As living organisms, microbes are susceptible to the conditions of the environment they inhabit. Individual species are typically only active over fairly narrow ranges of pH, redox potential and temperature. Microbes also require sufficient metabolites to produce energy and space in which to grow. Microbial activity may, therefore, be limited by:

- high pH environments (when present), although some micro-organisms are able to tolerate highly alkaline conditions.
- unfavourable redox conditions. Reducing conditions are unfavourable for bacteria introduced from the surface, but may encourage anaerobic bacterial activity.
- lack of metabolites. Although the complex chemistry of the near-field environment, particularly in the ILW/LLW disposal area, may provide sufficient metabolites for some microbial activity [199], the low permeability of some barrier materials, such as bentonite, may restrict the supply of nutrients. However, ILW wastes, particularly those containing organic materials such as cellulose and polymers, will undergo degradation in the GDF environment which will provide nutrients for metabolism of microbes [201].
- chemical processes, or the action of extracellular enzymes, which are essential precursors to significant biological activity.
- extremes of temperature, although some bacteria are capable of surviving even in the high temperature environment expected during the transient phase of a HLW and spent fuel disposal area.
- intense radiation, particularly close to the ILW, HLW and spent fuel, although the HLW/SF container may give a significant degree of attenuation [202].
- insufficient space, for example in a saturated bentonite buffer where the porosity is very low and pore spaces are typically smaller than typical cell diameters of microbes [199].

Despite the fairly extreme conditions in a GDF, it cannot be assumed that the near field will be a sterile environment. Although it is likely that sterile conditions will be found close to HLW and spent fuel, where temperatures will be highest, where there may be little water present, and where the radiation is most intense, there may however be numerous heterogeneously-distributed zones elsewhere in the near field where microbial activity will be viable. For example, microbes could inhabit the interfaces between different barriers.

Depending upon the specific disposal concept and the characteristics of the waste and other materials present, possible effects of microbial activity include:

- degradation of organic materials present in wastes and the subsequent generation of gases (as discussed above)
- microbially-influenced corrosion, for example, localised corrosion that arises as a result of the generation of aggressive species such as sulphides from microbial activity
- controlling the redox potential of environments relevant to the geological disposal of radioactive waste by iron(III)-reducing species [203]
- changes to the chemical form of some radioelements, such as uranium, technetium, neptunium and plutonium and for certain radionuclides such as carbon-14.

During the post-closure period the level of microbial activity in the near field will depend upon the prevailing conditions and on the nature of the waters and materials present. Microbial activity may fluctuate as the near field evolves, or may continue at steady levels over very long periods (see Task 443 in our Science and Technology Plan [10]).. There is, thus, uncertainty surrounding the implications of microbial activity in the near field of the GDF and this is likely to be a subject of ongoing research once site-specific information becomes available and concept development has progressed.

### 3.14 Container degradation

Many processes affecting the degradation of containers are coupled and interdependencies can be complex. Waste package longevity would be promoted by the selection of engineered barrier materials that will degrade only slowly and help to maintain conditions at container surfaces that keep corrosion rates low for very long periods.

A full description of the post-closure evolution of containers is given in the Waste Package Evolution status report [2]. The chemical conditions that will develop in the vicinity of the waste packages as the engineered barrier system evolves are a direct control on the rate of package evolution. These have been discussed throughout this report (for example, the evolution of pH and redox potential in cement-based and bentonite-based disposal systems).

Like many of the processes occurring during near-field evolution, many processes affecting containers are coupled and interdependencies can be complex. Some examples relating to container degradation that have already been discussed in previous sections of this report include:

- corrosion of metals and the generation of gas which can impact both the chemical and physical evolution of the engineered barrier system, and in particular the evolution of buffers and backfills (see subsection 3.11).
- corrosion of steel materials in the near field and the generation of iron corrosion
  products which can be considered to impact the sorption capacity of bentonite
  buffers. It is important to consider that iron-corrosion products, as well as being able
  to affect bentonite swelling properties, also have high sorption capacities in their
  own right and therefore will contribute to the containment of radionuclides in the
  engineered barrier system.

Typical corrosion mechanisms and rates in the GDF, and the factors that affect these, are well understood [204] and are discussed in more detail elsewhere [2]. In future, it will be important to evaluate the expected post-closure container corrosion mechanisms and rates for specific UK sites. When designing the engineered barrier system, waste package longevity will be promoted by the selection of engineered barrier materials that will degrade only slowly and that will help to maintain conditions at container surfaces that keep corrosion rates low for very long periods. For example:

- the illustrative concepts considered for the disposal of HLW and spent fuel in higher strength and lower strength sedimentary host rocks include a bentonite buffer to create a low permeability barrier around the waste container. This will inhibit advective flow and will keep container corrosion rates low for very long periods by limiting the transport of aggressive species such as sulphides and microbes to the container surface.
- in the illustrative concepts considered for the disposal of HLW/spent fuel and ILW/LLW in an evaporite host rock, the compaction and eventual sealing of the host rock and crushed salt buffer and backfills will minimise the availability of water for waste container corrosion. In this environment, in any case, containment is provided by the host rock rather than the container.
- In the illustrative concepts considered for the disposal of ILW/LLW in higher strength and lower strength sedimentary host rocks, corrosion of the waste containers is minimised by high pH and low Eh conditions in groundwaters, but the impacts of more saline waters also needs to be taken into account.

The Waste Package Evolution status report [2] describes the different corrosion processes that will affect waste packages and the degradation of the wasteforms and the release processes of radionuclides from them. Radionuclide behaviour in the near field is also discussed in the Behaviour of Radionuclides and Non-radiological Species in Groundwater status report [4].

### 3.15 Mechanical degradation of containers

In GDF concepts where the longevity of containment within the waste package is important, long-term mechanical evolution needs to be considered. For copper containers, two potential mechanisms for mechanical degradation are creep ductility and package shearing as a result of seismic strain.

In GDF concepts where emphasis is placed on the longevity of containment within the waste package, long-term mechanical and chemical evolution processes need to be considered. The principal example of this is the KBS-3 concept developed in Sweden and Finland, where the long-term integrity of the copper container is a key component of the safety case.

Three potential mechanisms that could lead to loss of integrity are currently receiving attention:

- creep ductility of copper
- failure from an isostatic load
- package shearing, as a result of seismic strain from fractures in the near-field rock.

Creep ductility over many hundreds or thousands of years is currently not a fully understood process and there is limited experimental or observational understanding. Accelerated tests at high strain rates might not reflect actual behaviour over the long term. Isostatic or distributed loads on the outer surface of the copper containers as a result of hydrostatic and lithostatic loads and the swelling of buffer material could cause slow creep of the copper as it is compressed onto the container internals, closing up small void spaces. If this creep were to occur and be uneven, it is feasible that the copper container might fail. If there is only slow resaturation of the buffer in some deposition holes (both SKB and Posiva estimate this could take up to some thousands of years in some locations in the GDF), this could also have an impact on copper creep. The creep behaviour of copper and welds in copper containers is summarised in references [205, 206].

This mechanism is important to safety case development as it is regarded as one of the only processes whereby 'common-cause' failure of many containers could occur, possibly at much earlier times than container failures due to corrosion. Consequently, copper creep will need to be understood better and we are monitoring overseas work in this area. Possible mitigation measures include the addition of phosphorous to the copper to improve its creep resistance.

Mechanical shear of containers as a result of seismicity has been studied most extensively for the case of copper containers emplaced in fractured, high-strength rocks in Sweden and Finland. Seismic events that might affect the EBS could occur in rock deformation zones in, close to, or deep beneath the GDF. This activity may arise as a result of the rock response to the heat load from the wastes or as a result of the specific stress circumstances involved when thick ice covers the GDF in an ice age and then melts. All of these circumstances are conceivable in the expected evolution of a GDF in the UK.

In higher strength rock, modelling of potential impacts has centred on the use of a rockmechanical modelling approach, whereby shear on a major deformation causes sympathetic shear in small ('target') fractures that could intersect a deposition hole [207, 208, 209]. The response of the container is a function of the strength and response of the fracture, the shear velocity on it and the state and behaviour of the buffer at the time the event occurs, and is strongly controlled by the strength and manufacturing quality of the container's internals. Based on the thickness of bentonite buffer specified in the KBS-3V design, SKB and Posiva require that shears at fractures cutting across deposition holes should not exceed 50 mm in order to preserve the integrity of the copper shell.

The approach taken by SKB and Posiva is to avoid locating container deposition holes where there are fractures of sufficient length to host shears of this magnitude [205]. Addressing this specific matter has become a control on GDF design and size. For example, a recent study by Posiva [210] indicates how applying the fracture avoidance criterion developed for the KBS-3V concept to the KBS-3H concept<sup>14</sup> could impact the spatial utility of the repository rock volume.

The 'classical' rock mechanics approach that has been applied to model the shear response of the fracture network in the GDF to seismic events is now being compared to alternative approaches. Recent work using the latter approach [211] suggests that even relatively small natural earthquakes that occur close to the GDF during the period when the rock is being heated by the waste could give rise to shears relevant to container integrity.

The mechanical behaviour of fractures and containers in response to a seismic event has to be linked to estimates of the likelihood and frequency of natural earthquakes of different magnitudes at different times. There are differing views on how best to derive a figure for the probabilistic assessment of container shear failure and it is important to ensure that estimates are sufficiently conservative. The appropriate frequency/magnitude values to use are highly dependent on the geological and regional/local tectonic environment of the GDF.

In future, analyses of seismicity and its impacts may need to be framed more strongly using seismological information about the nature of mid- to upper-crustal continental seismicity, in particular focal depths of earthquakes under present-day and post-glacial conditions and the upwards propagation of shear from mid to shallow depth, low magnitude events into GDF depths.

Even though existing analyses [129] indicate extremely low risks from earthquake shear of containers, the matter is ultimately highly site-specific. In addition, the GDF surface and operational facilities will be subject to conventional Probabilistic Seismic Hazard Analysis (PSHA) applied to all nuclear facilities and it may be appropriate to consider applying parallel techniques such as Probabilistic Fracture Displacement Analysis (PFDHA) to the GDF itself. All of these techniques would utilise broadly similar databases to those that have been used in the existing Swedish and Finnish studies.

### 3.16 Near-field evolution system understanding

As important as it is to demonstrate fundamental understanding for individual processes, system understanding considers how each of the engineered barriers will evolve through a combination of evolution processes.

The occurrence, rates and ultimate extent of the processes discussed above in subsections 3.4 to 3.15 will depend on the site characteristics and the disposal concepts selected. As important as it is to demonstrate fundamental understanding for each individual process, ultimately these processes will be considered collectively as a system.

System understanding considers how each of the engineered barriers will evolve through a combination of evolution processes. It includes understanding how these processes are

<sup>&</sup>lt;sup>14</sup> KBS-3H is an alternative concept developed by SKB with horizontally-emplaced containers.

coupled to each other and how they collectively impact on the engineered barrier system over various timeframes.

To demonstrate how understanding of individual processes can be used to develop system understanding, summaries of the expected near-field evolution specific to each of the illustrative disposal concepts used in the DSSC are provided in Section 4. Here consideration is given to the typical requirements on the respective engineered barrier systems imposed by the combination of waste types and the characteristics of the geological environment considered. This allows the choices for material selections to be understood for each of the engineered barrier systems. The summaries of evolution presented emphasise the important evolution processes and their significance for the illustrative disposal concept examples, explaining how the functions of their respective engineered barriers are fulfilled during the early post-closure and late post-closure timeframes.

In the future, our understanding of near-field evolution will need to consider the impacts of external influences on engineered barrier system performance in further detail. Such influences have been discussed throughout subsections 3.4 to 3.15 in a generic way. However only once site-specific characteristics are known and concept development has progressed, so that the detailed nature of the concept to be implemented and its design are known, will a full evaluation of these impacts be possible. In particular, construction, operational and closure activities (for example, waste emplacement and backfilling strategies which will influence the thermal evolution of the near-field and the rate of resaturation of disposal areas) will need to be considered.

In the current generic phase of our programme, our understanding of the processes discussed in subsections 3.4 to 3.15 will provide the basis for continuing to build our understanding of how an engineered barrier system will contribute to waste containment over the very long term (tens to hundreds of thousands of years following GDF closure). Such information is used as the basis to inform identification and development of GDF concepts, assessment of packaging solutions, the development of the Disposal System Technical Specification [14], and the on-going development of the Disposal System Safety Case.

To support these activities, uncertainties relating to near-field evolution processes continue to be actively researched through our research and development programme. Those research activities that we plan to undertake in the near-term to support our programme are detailed further in the Tasks numbered from 416 to 485 in our Science and Technology Plan [10].

## 4 Evolution of the engineered barrier system for illustrative geological concept examples

In this section, our understanding of near-field evolution based on the information available internationally for illustrative disposal concept examples is set out for three host rocks: higher strength host rocks (subsection 4.1), lower strength sedimentary host rocks (subsection 4.2), and evaporite rock (subsection 4.3).

In the subsections that follow, the discussion is limited to the evolution of the engineered barrier system, making reference where appropriate to the other status reports. In particular, the fate of individual radionuclides in the engineered barrier system is not considered – this is discussed in the Behaviour of Radionuclides and Non-radiological Species in Groundwater status report. In order to evaluate the performance of the system as a whole, the Environmental Safety Case [20] considers the evolution of the engineered barrier system *in conjunction* with other barriers and the fate of radionuclides throughout, and thereby the implications for safety – so while there is some overlap in the parts of the Environmental Safety Case [20] that discuss the evolution of the engineered barrier system, that report and this one are not providing the same narrative. Specifically, this report describes our understanding but does not attempt to make safety related judgements about the evolution of the engineered barrier system or to discuss conservative assumptions made in the safety case [20].

## 4.1 Evolution of the engineered barrier system in a higher strength host rock

Concepts developed in Sweden for spent fuel disposal and in the UK for disposal of ILW are used as illustrative concepts to demonstrate our understanding of engineered barrier system evolution in a higher strength host rock.

Subsection 4.1.1 describes the typical characteristics of a higher strength rock. The typical requirements on the engineered barrier system are described in subsection 4.1.2. The key features of these illustrative concepts are described in subsections 4.1.3 and 4.1.5, and the expected evolution of the engineered barriers for these concepts is discussed in subsections 4.1.4 and 4.1.6.

### 4.1.1 Typical characteristics of a higher strength host rock

Underground construction is relatively straightforward in higher strength rocks, which typically have low matrix permeability, but a network of faults and fractures may be present.

Higher strength host rocks provide a high degree of mechanical strength and stability, which enables relatively straightforward underground construction and long-term stability of large openings/caverns without the need for extensive rock support. This allows great flexibility with respect to GDF design and accommodation of different waste packages, as well as facilitating staged operation of a GDF and/or delayed backfilling of excavated regions, if required. It should be possible to construct large caverns (16m span or more) that are intrinsically stable and require only limited reinforcement of the host rock at depths of up to 1000m [212, 20].

Higher strength host rocks typically comprise igneous or metamorphic rocks such as granite, or geologically older sedimentary rocks. The matrix of these rocks typically has a low permeability. However, due to their geological history (which may include magma cooling, hydrothermal events and/or tectonic movements), a network of faults, fractures,

fissures and/or shear zones may be present within this type of geological environment [20]. This may allow advective flow of groundwater through the host rock, and hence, relatively rapid transport pathways may exist, compared to those in lower strength sedimentary rocks or evaporite rocks. The presence of these features means that the evolution of the engineered barrier system may be spatially and temporally heterogeneous. It may be desirable to artificially seal some fluid-bearing features. Various silica sol gels and cement formulations have been considered for this purpose [213], but it will be important that the use of fracture grouts during construction does not impinge upon the long-term performance of the GDF.

The magnitude of the EDZ created in this geological environment may be relatively small (depending on factors such as the excavation technique). However, once created, sealing and recovery of the EDZ will be relatively slow, since processes such as rock creep will be minimal. The EDZ may therefore have a more significant, prolonged effect on near-field evolution than in other geological environments [3].

The relatively low solubility of minerals present in higher strength host rocks, coupled with relatively rapid advective transport in environments where the fracture network is well connected from surface to depth, may lead to groundwaters with a relatively low salinity in the upper hundreds of metres of rock. Such a chemical environment would have a favourable effect on the corrosion rate of metal components of an engineered barrier system employed [2]. However, at greater depths, or in less connected systems, or in environments where the higher strength rocks are hydrogeologically isolated by a thick sequence of overlying sediments, highly saline conditions can prevail. Whilst there is the potential for increased corrosion, such conditions could indicate hydrogeological isolation; a highly favourable hydrogeological characteristic.

Higher strength host rocks typically have relatively low thermal conductivity. Consequently, peak near-field temperatures may be higher than in other, more thermally-conductive geological environments. Thermal effects are likely to be less spatially extensive, but may persist in the host rock for longer timescales.

### 4.1.2 Typical requirements on the near field in a higher strength host rock

Long-term containment in a higher strength host rock can be achieved by a durable engineered barrier providing physical containment, or a chemical barrier that reduces the mobility of radionuclides.

Given the relatively high permeability that may be a feature of such geological environments if they are considerably fractured, disposal concepts developed for application in a higher strength host rock typically incorporate an engineered barrier system designed to provide a high degree of containment over a long timescale. Such an approach complements the isolation provided by the geosphere. Depending on the inventory and the nature of the waste for disposal, long-term engineered containment can be achieved in two ways:

- by employing a highly durable, long-lived engineered barrier system that provides extensive physical containment, such as a thick-walled or high integrity container
- by implementing an engineered barrier system that provides substantial chemical containment and hence, reduces the mobility of radionuclides in the near field.

The two illustrative concepts discussed in this subsection are each designed to fulfil one of the alternative approaches to containment described above.

## 4.1.3 Illustrative concept for disposal of HLW and spent fuel in a higher strength host rock

In this concept, spent fuel in a copper container is placed in deposition holes and surrounded by compacted bentonite buffer. Under these conditions the container is anticipated to provide containment for in excess of 100,000 years, during which time a very large proportion of the radionuclide inventory will typically have decayed.

The Swedish KBS-3V disposal concept is illustrated in Figure 18. In this concept, spent fuel is placed in a high integrity copper container. This is supported by a cast iron insert which provides mechanical strength. The container is then placed directly in deposition holes excavated in the rock and surrounded by a low permeability compacted bentonite buffer [29, 214]. Access tunnels are then backfilled with bentonite [30]. The container is capable of providing containment over a long period, potentially 100,000 years or more [50, 51]. The longevity of the container is underpinned by the use of corrosion-resistant materials and the provision of a near-field environment in which corrosion and associated processes occur extremely slowly. We recently carried out a review of the KBS-3V concept, focussing on the development of bentonite barriers [215].

## Figure 18 The KBS-3V concept for the disposal of spent fuel in a higher strength host rock in Sweden [29].



Figure 19 illustrates the isolation and containment functions provided by the engineered barriers of the KBS-3V concept together with the geosphere.

In this concept, the primary function of the engineered barriers is to provide complete containment of the waste for many tens of thousands of years, and thereafter, to retard the release of radionuclides from the engineered barrier system. In this way, the engineered barrier system will complement the containment and isolation provided by the geological barrier. Emphasis is placed on the containment provided by the copper container in concert with the bentonite buffer [29].

# Figure 19 The multiple barriers present in the illustrative concept (adapted for disposal of UK HLW and spent fuel) in a higher strength host rock.



The engineered barriers included in this disposal concept are described below:

**Wasteform:** The wasteform includes cylindrical pellets of uranium dioxide stacked in cladding tubes of zircaloy, a durable zirconium alloy, or stainless steel. The uranium dioxide wasteform will provide a stable, low solubility ceramic matrix that limits the release of the majority of radionuclides by dissolving slowly in groundwaters that come into contact with it [14].

**Container:** The highly corrosion resistant copper container will provide containment for an extensive period. By the time the container is breached, substantial radioactive decay will have occurred.

The cast iron insert provides mechanical strength. This is required in order to:

- withstand the combined external overpressure exerted by the rocks overlying the GDF, and the hydraulic pressure
- withstand loads associated with shear displacements in the rock associated with earthquakes
- withstand loads from bentonite swelling, which may be heterogeneous and which could otherwise cause crushing and/or bending of the container.

The internal structure of the insert will also maintain separation of individual fuel assemblies and, as the container remains intact for many tens of thousands of years with no water ingress, the likelihood of criticality is extremely small [6, 216].

**Buffer:** The bentonite buffer swells as it resaturates and through this process provides a low permeability barrier which minimises fluid flow and ensures that solute transport is by diffusion only. This limits the presence of water and aggressive species at the container surface. The high pore pressure in the bentonite also acts to limit microbial activity that could otherwise accelerate corrosion, and the high density bentonite filters colloids so that they do not reach the container surface in high quantities. The buffer also has a buffering effect on the pH and the redox potential of the near field, and conditions the porewater that contacts the container surface such that corrosion is limited.

The buffer holds the heavy container securely in its disposal position. It absorbs shear displacements in the rock, and so protects the container from disruptive events such as earthquakes. In the KBS-3V concept, the compaction of the bentonite is set to achieve a specific final density in the water-saturated buffer. The density requirement for the saturated buffer in this design is 1950 to 2050 kg m<sup>-3</sup> [29].

Once the container is breached, the buffer slows the release of radionuclides from the near field by providing an impermeable barrier that inhibits advective transport, and by providing a high surface area for sorption of radionuclides. The high cation exchange capability of bentonite also encourages radionuclide sorption.

**Backfill:** The backfill fills the tunnels above the disposal boreholes, and provides sufficient confining pressure to allow the bentonite buffer to swell sufficiently without escaping the disposal boreholes, so that it can function correctly. It is important that the backfill is emplaced soon after container/buffer emplacement, to prevent detrimental buffer expansion. The backfill also provides a barrier to human intrusion and limits advective transport through the near field.

# 4.1.4 Summary of evolution of the engineered barrier system for the illustrative concept example for disposal of HLW and spent fuel in a higher strength host rock

Once resaturated, the bentonite buffer provides a low permeability barrier that protects the container by limiting the transport to the container surface of species that might promote corrosion. The vast majority of the copper containers are expected to retain their physical integrity for a very long time.

This subsection provides a summary of the key steps in the near-field evolution of the illustrative concept for disposal of HLW and SF in a higher strength host rock. The discussion draws on the description of expected evolution set out by SKB for the undisturbed performance of the KBS-3V concept [29, 30]. We have also drawn on the equivalent Posiva safety case [129] and understanding of the behaviour of this disposal concept, based on the work completed previously by Nirex, and more recently by NDA RWMD [217, 218].

The approximate timescales over which key processes dominating the near-field evolution of this illustrative concept are expected to occur are illustrated in Figure 20. Many of the processes identified (such as container degradation, gas generation and buffer alteration) are expected to occur very slowly over many thousands or tens of thousands of years. These processes can be described independently, but some are coupled to each other and need to be considered within the context of the disposal system.

# Figure 20 Timeframes over which key near-field processes are expected to occur for the illustrative concept for disposal of HLW/SF in a higher strength host rock.



T=0 is closure of the GDF.

#### Evolution during construction and operation of the GDF

An EDZ will be created as soon as excavation activities commence, due partly to excavation-related vibrations and partly to changes in the stress fields in the rock, which lead to brittle fracturing. The EDZ will have different mechanical and hydraulic properties to the intact host rock; it may be more extensively fractured, and consequently, would be more permeable, as well as exhibiting a lower fluid pressure in pores and fractures. Depending on the excavation technique and the nature of the site-specific geology, it may extend up to several tens of centimetres from the excavated region [219]. The redistribution of stress may cause rock spalling to occur and/or the reactivation of pre-existing fractures in the host rock.

The pre-existing hydraulic gradients present in a given geological environment will be disrupted by excavation of the GDF. Some fluid is likely to migrate along hydraulic gradients towards the engineered barrier system until the hydraulic potential of the GDF is restored to that of the surrounding host rock. Changes in the saturation of the host rock in response to changes in the hydraulic gradient are likely to occur relatively quickly, particularly if the host rock is significantly fractured.

Depending on the permeability of the rock and the GDF design, the chemistry of groundwater entering the engineered barriers (particularly the pH, redox potential and/or salinity) could potentially change as a result of infiltration of near-surface waters into deeper parts of the bedrock and/or upwelling of more saline waters from depth. Aerobic conditions will predominate in the near field, potentially due the to ingress of near-surface waters, and because the facility will be ventilated to the surface. Minor thermal perturbations of the host rock surrounding the excavated spaces may also occur, due to ventilation of these areas.

Although, as discussed earlier, the EDZ may be able to seal through processes such as mineralisation in fractures, there is unlikely to be significant sealing through rock creep, since higher strength rocks tend to be quite rigid. As a result, any recovery of the EDZ is likely to be slow. To counteract this, consideration can be given to sealing any significant EDZ fractures in disposal regions of the GDF (which might otherwise provide a rapid transport pathway), by filling them with some form of grout. If used, any cementitious grout materials may begin to react with groundwater and other components of the near field, leading to increased pH of water in the near field, and potentially affecting the presence of colloidal species and/or the sorption properties of the backfill, buffer and surrounding host rock. The onset of such interactions may be relatively quick, once the grout is emplaced.

As soon as the bentonite buffer and the tunnel backfill have been emplaced, the bentonite will begin to swell, as it absorbs moisture from humidity in the air and any groundwater it comes into contact with. Eventually the bentonite buffer will swell to the point where it completely seals the space between the waste package and the host rock and forms a uniform low permeability barrier encapsulating the disposal container. Prompt backfilling and sealing of the disposal tunnels is essential to ensuring that the saturated density of the bentonite buffer is sufficiently high that it can fulfil its required functions and to reduce the period over which piping erosion might occur. The methodology for emplacement of the engineered barriers within the near field is therefore of key importance in ensuring that the buffer attains a sufficiently high density to protect the container from degradation and is an area where further testing and demonstration work is planned in Sweden and Finland. The time taken to fully resaturate an individual deposition hole in a higher strength rock could vary considerably between deposition holes, from a decade or so to several thousands of years, depending on the characteristics and spatial variability of the fracture network, which control inflows to individual deposition holes. Resaturation of the backfill in the disposal tunnels would take decades to centuries. Thus, the early deposition holes, and possibly some of the early disposal tunnels, could be fully resaturated before final closure of the GDF, while other deposition holes might not resaturate completely until after the postclosure thermal period.

The waste will initially give off significant quantities of heat as a result of decay of the shortlived component of the inventory. While the disposal tunnels are open, we will use the ventilation system to ensure that temperatures in the open parts of the GDF remain within specified limits. Once a disposal tunnel has been sealed, the temperature in the buffer and in the surrounding rock may rise several tens of degrees above ambient. Thermal constraints to a specified temperature limit are a significant factor determining package spacing, underground layout and the optimum times at which to remove waste packages with different thermal outputs from storage and emplace them in the GDF.

The final stage of the closure engineering is the backfilling of the various access tunnels and shafts and emplacement of the main low-permeability seals. Closure completes the isolation of wastes and aims to assist in restoring the natural flow and chemical conditions as well as is possible. The intention is to ensure that the sealed openings do not constitute a preferential hydraulic or gas pathway connecting the surface and deep environments. For example, ideally, the permeability of the seals should be no higher than the rock in which they are emplaced. Closure also acts as an impediment to intrusion by people in the future.

During this early period of GDF evolution our understanding is that the waste packages, which are protected by the bentonite buffer, will provide complete containment of the waste.

#### Early post-closure evolution

The resaturation process described above will continue to completion. The bentonite buffer provides a low permeability barrier around the disposal container that protects the container by limiting the rate of transport to the container surface of water and aggressive

species such as sulphides and microbes that might promote corrosion and then by limiting the rate of transport of any corrosion products away from the container. The buffer also protects the container from events such as possible seismically induced shear movements on small discontinuities that might intersect the deposition hole.

During this period, the waste gives off significant quantities of heat until the short-lived component of the radionuclide inventory has decayed. As a result, the temperature of the buffer and the surrounding rock may rise to several tens of degrees above ambient before slowly decreasing. The peak temperature in the engineered barrier system is likely to occur within a few decades of waste emplacement, but we expect temperatures to remain above ambient for up to 10,000 years. The elevated temperatures will affect the solubility of minerals in the bentonite, and hence the porewater pH and mineralogy, leading to spatial variation in porewater characteristics, depending on the thermal regime. The temperature increase and the effects of irradiation from the waste may also have other effects on the chemical and mechanical stability of near-field components. However, the copper container is expected to remain intact for at least 100,000 years. By the time significant container degradation has occurred, a high proportion of the radionuclide inventory is expected to have decayed.

In addition to the swelling that occurs on resaturation, interaction with the host rock groundwater will result in some minor changes to the physical and chemical properties of the bentonite buffer, depending on the groundwater chemistry (for example, its salinity). Some minerals may dissolve and others may precipitate; ion-exchange reactions will also occur. These reactions will result in the development of reducing conditions and will buffer the bentonite pore water to near-neutral to slightly alkaline pH. The reactions also have the potential to influence the swelling pressure of the bentonite. Elevated temperatures may increase the rates of some of these reactions and may influence fluid movements, in particular the transport of water vapour, which is an important process in the resaturating bentonite.

The chemical conditions in the surrounding host rock will slowly return to values close to undisturbed conditions over tens of thousands of years, depending on the transport properties of this region. The groundwater flow field will slowly recover to equilibrium with the surrounding environment once the near-field rock and the rock overlying the GDF are fully resaturated and natural hydraulic gradients are re-established.

#### Late post-closure evolution

It is conservatively assumed that a small number of the copper containers, those with undetected manufacturing defects, may cease to provide complete containment from about 10,000 years onwards. However, the vast majority of the copper containers are expected to have a lifetime of at least 100,000 years and probably much longer. Once a copper container has failed, water would access the cast iron insert, which would corrode, generating hydrogen gas [29]. If the rate of gas production exceeds the rate at which gas can be transported away by diffusion, a free gas phase could form and the pressure increase could contribute to growth in the container penetration hole.

The wasteform would start to dissolve in the water entering the container and concurrently would release radionuclides that have not decayed *in situ* [2, 4, 29]. In the case of HLW (not part of the Swedish waste inventory) the majority of the radionuclide inventory will have decayed by this time; slow dissolution of the glass matrix will limit the release rate of any remaining radionuclide inventory. For spent fuel, the portion of the instant-release

fraction<sup>15</sup> that has not decayed *in situ* would be released rapidly; the spent fuel matrix would then dissolve slowly and release the remaining radionuclides [2,4,29]. The bentonite buffer is expected to provide an effective barrier to the transport of radionuclides released from the fuel once the container has been penetrated. Any radionuclides that are released from the near field would be retarded, and potentially contained, in the geosphere.

In the very long term (many hundreds of thousands to millions of years) the engineered barrier system is not expected to provide full containment, although some of the copper containers may remain intact. However, the degraded barriers are likely to continue to retard the release of contaminants to some degree, although by this time the vast majority of the radionuclide inventory will have decayed.

## 4.1.5 Illustrative concept for disposal of ILW and LLW in a higher strength host rock

In this concept, ILW and LLW are encapsulated in cementitious grout in stainless steel containers and placed in disposal modules. These are backfilled with a cementitious material designed to provide a long-term chemical barrier.

The illustrative concept example for disposal of ILW/LLW in a higher strength host rock is based on the UK Phased Geological Repository Concept (PGRC), previously developed by Nirex for a higher strength, low permeability rock [178, 179]. This disposal concept is illustrated in Figure 21. In this concept, ILW and LLW, typically encapsulated in a cementitious grout<sup>16</sup> in stainless steel containers<sup>17</sup> will be emplaced in disposal modules excavated in the rock. The use of rock bolts, metal mesh and shotcrete is envisaged to provide engineered support to the excavations. Concrete linings are likely to be required in the access tunnels [212]. At some point the disposal modules will be backfilled with a cementitious material, such as Nirex Reference Vault Backfill (NRVB), designed to provide a chemical barrier over the long term. Access tunnels will also be backfilled with cementitious material and low permeability seals.

<sup>&</sup>lt;sup>15</sup> The portion of the inventory (fission and neutron activation products) that has segregated to the grain boundaries in the wasteform and may be released rapidly upon exposure to groundwater.

<sup>&</sup>lt;sup>16</sup> Alternative conditioning methods such as polymer encapsulation or vitrification are under consideration for some wastes [2].

<sup>&</sup>lt;sup>17</sup> Alternative container materials such as ductile steel and concrete are under consideration for some wastes.

# Figure 21 Illustrative concept example for ILW and LLW in a higher strength host rock.



Figure 21 illustrates the engineered barriers and natural barriers present in this illustrative disposal concept example. In this concept, the cementitious backfill is designed specifically to create and sustain an alkaline environment in which the solubility of many key radionuclides will be reduced. It also has a high porosity, presenting a large surface area available for sorption of radionuclides. The engineered barrier system will complement the containment and isolation provided by the geological barrier and will ensure long-term safety. Emphasis is placed on the chemical containment provided by cementitious wasteforms and the buffer/backfill.

The engineered barriers included in this disposal concept are described below:

**Wasteform:** The wasteform will provide a stable, low solubility matrix that limits the release of the majority of radionuclides by dissolving slowly in groundwaters that come into contact with it [14].

**Container:** The steel container will provide a degree of mechanical strength (depending on the relative thickness of the container being considered). Steel will slowly corrode in a way that will aid the maintenance of chemically reducing conditions (as will much of the metal present in the waste). Most containers have vents to allow the release of gases, avoiding gas over-pressurisation damaging the waste packages. Corrosion products will also provide a high sorption capacity for radionuclides in the long term.

**Buffer/backfill**: A cementitious buffer such as NRVB will create relatively uniform, alkaline chemical conditions. These conditions will significantly reduce the solubility of many radionuclides dissolved in the groundwater within and around the near-field. High pH conditions also reduce metal corrosion rates and suppress microbial activity, thus supporting the longevity of the container. The cement-based backfill also has a high capacity for sorbing radionuclides, thus inhibiting the migration of radionuclides away from the engineered barrier system.

**Mass backfill:** The access tunnels will be backfilled with a cementitious material, or most probably with crushed host rock. Mass backfill would provide mechanical support to

prevent the voids from collapsing as the host rock evolves and any engineered support degrades, thereby mitigating the potential for further damage to the geosphere. The low permeability of the backfill will prevent the access tunnels from acting as preferential pathways for fluid migration. The backfill will also provide a barrier to human intrusion.

**Tunnel seals:** Individual disposal tunnels will be sealed and then the access tunnels will be backfilled. A combination of higher permeability seals using crushed rock combined with low permeability seals using concrete dams is assumed [212, 220]. These seals will act to isolate the disposed wastes from the rest of the disposal system and to prevent access tunnels, drifts and shafts from acting as preferential pathways for groundwater flow and radionuclide transport.

# 4.1.6 Summary of the evolution of the engineered barrier system for the illustrative concept example for disposal of ILW and LLW in a higher strength host rock

On resaturation, alkaline conditions will become established, limiting the solubility of many radionuclides. The alkaline conditions will persist for a long time (tens to hundreds of thousands of years) before the backfill becomes altered through reaction with groundwater and loses some of its effectiveness.

This subsection provides a summary of the key steps in the near-field evolution of the illustrative concept for disposal of ILW and LLW in a higher strength host rock. The discussion draws in particular on work undertaken in the UK over the past 30 years to develop a geological disposal concept for the disposal of ILW.

The approximate timescales over which key processes dominating the near-field evolution of this illustrative concept are expected to occur are illustrated in Figure 22. Many of the processes identified (for example, cement evolution, and gas generation) are expected to occur very slowly over many thousands or tens of thousands of years. These processes can be described independently, but they are strongly coupled to each other.

# Figure 22 Timeframes over which key near-field processes are expected to occur for the illustrative concept for disposal of ILW and LLW in a higher strength host rock.



T=0 is closure of the GDF.

#### Evolution during construction and operation of the GDF

An EDZ will be created as soon as excavation activities commence, due partly to excavation-related vibrations and partly to changes in the stress fields in the rock, which lead to brittle fracturing. Evolution of the EDZ will be fairly similar to that described above for the illustrative HLW and spent fuel concept in a higher strength host rock. For ILW/LLW regions of the GDF, the use of cement-based fracture grouts and rock support systems is compatible with the cementitious backfill and buffer considered and any high-pH plumes arising from fracture grouting are likely to be minor in comparison to the impact of chemical conditions provided by any backfill.

During the operational period, the ILW/LLW vaults and the access tunnels will be maintained in a dry and ventilated state. This will result in some desaturation of the host rock around the excavations. The rock immediately adjacent to the excavations may also undergo chemical changes as a result of oxidation reactions or as a result of reactions with materials, most likely concretes, that are used to provide structural support to the excavations. Some of the ensuing reactions may contribute to the sealing of fractures in the EDZ referred to above. The presence of a GDF that is maintained in an open and ventilated state for many decades could, through the flow it induces, potentially lead to changes in the composition of the groundwater in the surrounding host rock. For example, upconing of more saline water from depth is considered to be a possibility in Scandinavia [29]. Minor thermal perturbations of the host rock surrounding the excavated spaces may also occur.

The ILW/LLW packages will be in an aerobic environment throughout the operational period. Under these conditions it is expected that the degradation processes (primarily corrosion processes) will be different to, and in some cases more rapid than, those expected to dominate in the long term, after closure. The Waste Package Evolution status report [2] describes the various degradation processes that might affect waste packages, their likely rates and the environmental and other parameters that control them. Through

appropriate design of the operational regime it is possible to ensure that waste package degradation during the operational period is kept to a minimum, for example by ensuring that groundwater cannot come into contact with the packages and that temperature and humidity are controlled within pre-defined limits. Gas will be generated within some of the waste packages [5], and any that is released through the vents will be removed by the ventilation system and discharged in accordance with the GDF's discharge authorisation [5, 221, 222]. Similarly, water pumped from the vaults will be monitored and treated if necessary in the effluent management system, before discharge [221].

The ILW/LLW disposal vaults will be backfilled with NRVB as part of the phased closure of the GDF. Emplacement of the NRVB will transiently increase the temperature and will bring water into contact with the waste packages. This is likely to result in the onset of general corrosion of the waste packages, and hence an increase in the gas generation rate [22], although the rate will be low because the chemistry of the water will be conditioned by cement. Delaying backfilling until closure extends the period of reversibility with little effort.

#### Early post-closure evolution

Following closure, the ILW/LLW disposal area will start to resaturate. The engineered barrier system will cease to provide full physical containment shortly after resaturation because waste containers are vented. The time taken for the ILW/LLW disposal vaults to resaturate fully will depend on the properties of the host rock, but in a typical higher strength host rock it is expected to be of the order of a few decades to a few centuries [22]. The incoming groundwater would rapidly equilibrate with the NRVB, resulting in the development of alkaline conditions. Corrosion reactions will result in the establishment of reducing conditions.

Although high pH and reducing conditions will limit the rate of corrosion [2], gas will be generated by the corrosion of Magnox, aluminium and steel in the waste and other engineered components, and by the degradation of organic materials. The gas composition will be predominantly hydrogen, with some carbon dioxide and methane [5, 22]. While some of the gas will dissolve in the groundwater it is likely that a free gas phase will form. However, it is expected that gas will be able to escape relatively easily into the host rock [5], although its presence may slightly reduce the rate of resaturation. Depending on the gas transport properties of the geosphere at the site, some of this free gas might reach the surface [223].

Once the disposal vault saturates, small amounts of the ILW/LLW inventory will begin to be released, in dissolved form, through the vents in the ILW containers into the backfill. It is expected that the quantity of cementitious backfill used in the ILW/LLW disposal area will be sufficient to maintain alkaline reducing conditions in the ILW/LLW disposal area for at least a hundred thousand years. These conditions limit the rate of corrosion and the activity of microbes that degrade the organic components of the waste, and hence limit the gas generation rate. The high pH also limits the solubility of many radionuclides and non-radioactive species, while the minerals in the cement and the backfill (which have a high available surface area) promote sorption of key radionuclides [4, 22]. The wasteform and the chemical barrier provided by the backfill are both important barriers limiting the rate of release of contaminants from the near field. Nevertheless, some radionuclides, particularly those in anionic form (such as <sup>129</sup>I and <sup>36</sup>CI) will be less retarded than the bulk of the inventory.

During this period, the engineered barriers are degrading very slowly as a result of interaction with the groundwater and with each other. However they will remain sufficiently intact that they are able to perform their intended functions fully. Throughout this period, the near-field porewater would be conditioned to high pH by the dissolution of components of the near-field cementitious materials. Cracking of both the backfill and the wasteforms is likely [2, 22], although it is not expected to have a significant impact on the overall

performance of the cementitious barriers. The potential impact of organic complexing agents derived from the wastes, which can increase radionuclide mobility [4], will become progressively less significant as their concentration in the waste is reduced by chemical, radiolytic and microbial degradation, as well as by processes such as dilution or sorption [4].

An alkaline disturbed zone is expected to develop around the ILW/LLW disposal area as a result of reactions between the host rock and porewater that has been chemically conditioned by the cementitious backfill. The resulting mineral dissolution and precipitation will alter the hydrogeological properties of the host rock around the GDF. It is expected that there will be a net decrease in porosity and permeability as fractures, especially those in the EDZ, become filled with new, relatively high volume, minerals.

### Late post-closure evolution

Many hundreds of thousands of years after closure, the engineered barriers in the ILW/LLW disposal area will begin to become less effective and will no longer be able to perform their intended functions fully. With time, the wasteforms and the NRVB will be altered through reaction with the groundwater and eventually the pH in the ILW/LLW disposal area will fall to a level where the chemical barrier provided by the NRVB loses some of its effectiveness. However, sufficient NRVB will be included in the GDF design to take account of such reactions so as to maintain the chemical barrier for the required timescale. Those radionuclides that have not decayed within the waste packages and near field may be released to the host rock.

Gas will continue to be produced from the corrosion of metals, but the production rate is expected to have significantly declined by this time [5].

In the very long term (many hundreds of thousands to millions of years) the engineered barrier system is not expected to provide complete containment. However, the degraded barriers are likely to continue to retard the release of contaminants to some degree by chemical containment, although by this time the vast majority of the inventory will have decayed.

## 4.2 Evolution of the engineered barrier system in a lower strength sedimentary host rock

Disposal concepts developed in Switzerland for HLW and spent fuel, and for ILW disposal in the Opalinus Clay, are used as the illustrative disposal concept examples to demonstrate our understanding of near-field evolution in lower strength sedimentary host rock.

Subsection 4.2.1 describes the typical characteristics of a lower strength sedimentary rock. The typical requirements on the engineered barrier system are described in subsection 4.2.2. Both illustrative disposal concept examples (as illustrated in Figure 23) place an emphasis on the presence of a durable engineered barrier system. It should however be noted that the concept examples are defined by the properties of the Opalinus Clay and other lower strength sedimentary rocks might show some differences - particularly in the degree of creep closure as this is particularly dependent on the extent of induration (hardening) of the rock. The key features of these illustrative concepts are described in subsections 4.2.3 and 4.2.5, and the expected evolution of the engineered barriers for these concepts is discussed in subsections 4.2.4 and 4.2.6.

## Figure 23 Nagra's proposed layout for a deep geological disposal facility in Opalinus Clay, diagram courtesy of Nagra [33].



## 4.2.1 Typical characteristics of a lower strength sedimentary host rock

Lower strength sedimentary rocks are typically rich in clay materials and have low permeability. Fractures are largely absent since the rock self-heals though creep. Solute transport tends to be diffusion-dominated and therefore slow.

Lower strength sedimentary rocks are fine-grained, sedimentary rocks with a high content of clay minerals that provides their low permeability and are mechanically weak, so that open fractures cannot be sustained. Examples include clay and mudstone-dominated formations.

The characteristics of lower strength sedimentary rocks vary considerably, depending on the mineralogical composition of the rock, its depth and burial history, the temperature and the quantity/composition of porewater present. Poorly indurated lower strength sedimentary rocks (such as the Boom Clay under consideration for disposal of HLW in Belgium) typically have low mechanical strength and can creep (flow plastically) at relatively fast rates. This often means that fractures are largely absent, since the rock self-seals through creep closure. More highly indurated lower strength sedimentary rocks (such as the Opalinus Clay in Switzerland and the Callovo-Oxfordian clay in France) tend to be more brittle, and exhibit more extensive fracturing as a result; they are also less able to self-seal through creep. Generally, the plasticity of the rock tends to increase as the proportion of clay minerals rises.

Lower strength sedimentary rocks typically have low permeability, and fluid migration or solute transport tends to be diffusion-dominated, and therefore slow. Any fractures present in clay-rich mudrocks may be effectively sealed through development of clay smears along planes of movement. Groundwater flow velocities are therefore expected to be very low. This means that a lower strength sedimentary host rock typically provides a relatively stable chemical environment. It also means that the disturbances associated with construction and operations of the GDF are expected to be relatively localised (to within a

tunnel width or so). Many radionuclides, including actinides, sorb strongly to clay minerals, which enhances the containment provided by lower strength sedimentary rocks.

Lower strength sedimentary rocks also tend to limit the movement of gas [144]; indeed they form the cap rocks in many hydrocarbon deposits. Gas generated within disposal areas may not be able to migrate out of the engineered barrier system as a free gas phase. In the case of some lower strength sedimentary rocks (for example, clay), the rates of gas generation may be limited by the supply of water from the host rock to the GDF. A free gas phase would form, but because clay has very small inter-granular pores, the gas would find it difficult to migrate away from the GDF. The pressure would increase, leading to migration through porosity dilation and localised micro-fissuring in unfractured clays and mudrocks and finally, if the pressure were to continue to increase and exceed the mechanical stress field, fracturing [5]. For the lower strength sedimentary host rock environment (Opalinus Clay) being investigated by Nagra, the gas should be able to enter the EDZ in the host rock at pressures below those required for macroscopic fracturing.

## 4.2.2 Typical requirements on the near field in a lower strength sedimentary host rock

Lower strength sedimentary rocks provide substantial containment of radionuclides. Low mechanical strength limits the size of excavations and engineered barrier system components must have sufficient strength to maintain their functions when affected by convergence.

The low permeability, geochemical stability and high sorption capacity of lower strength sedimentary host rocks means that they provide substantial containment of radionuclides in their own right. Disposal concepts that have been developed for application in this type of rock often place an emphasis on the host rock to demonstrate long-term safety.

The low permeability of a lower strength sedimentary host rock may be affected by the heat produced by some wastes, which affects the locality of the GDF (for example the EDZ). Therefore, the engineered barrier system is specifically designed to provide complete containment for the duration of the thermal phase (whilst the waste is significantly heat-producing) or longer, which satisfies one of the requirements of the IAEA safety standards for radioactive waste disposal [21].

Some form of waste package will be required to facilitate waste emplacement and handling during the operational phase. This could also provide physical containment during the early post-closure phase. Other engineered barriers could also be employed to provide additional physical and/or chemical containment. Typically, emphasis is placed on buffer materials.

The mechanical properties of this host rock place further requirements on the design of the engineered barrier system:

- the relatively low mechanical strength of these rocks limits the size of excavated spaces, and disposal concepts for application in such geological environments often adopt a tunnel-based, or small vault-based layout rather than employing large span vaults. Engineered supports such as rock bolts and tunnel linings are also likely to be required to keep excavations open during the operational phase.
- some convergence of excavations is expected during the post-closure period as the host rock creeps, and as engineering supports degrade and eventually fail. The engineered barrier system components must have sufficient mechanical strength to maintain their integrity and their functions when affected by such convergence. But it is also important that construction materials do not degrade the host rock.

## 4.2.3 Illustrative concept for disposal of HLW and spent fuel in a lower strength sedimentary host rock

In this concept, HLW and spent fuel are placed in tunnels in thick-walled carbon steel containers, located on compacted bentonite blocks, surrounded by bentonite granules.

In this concept, HLW and spent fuel are placed in thick-walled carbon steel containers. In the case of spent fuel, the inside of the container is machined with a square, grid-like internal support for spent fuel assemblies (see Figure 24).



Figure 24 Spent fuel disposal in a thick walled carbon steel container.

For HLW, stainless steel flasks containing vitrified HLW are placed directly into the carbon steel container. The containers are then placed on compacted bentonite blocks in horizontal disposal tunnels excavated into the host rock, and surrounded by compacted bentonite granules. No other backfill material is used. The disposal tunnels will be closed with substantial seals to resist the swelling pressure of the bentonite buffer as it resaturates, immediately after completion of waste emplacement [33]. The disposal tunnels are not lined, but some form of concrete or shotcrete lining may be employed to provide structural support in access tunnels.

Figure 23 illustrates the engineered and natural barriers present in this illustrative disposal concept example. The primary function of the engineered barriers is to provide complete containment of the waste during the period of heat generation and beyond. In this way, the engineered barrier system will complement the containment and isolation provided by the geological barrier and ensure long-term safety. Emphasis is placed on the containment provided by the carbon steel container and the bentonite buffer [33].

The engineered barriers included in this disposal concept are described below:

**Wasteform:** The wasteform will provide a stable, low solubility matrix that limits the release of the majority of radionuclides by dissolving slowly in groundwaters that come into contact with it [14].

**Container:** The thick carbon steel container is expected to provide containment over a 10,000 year lifetime (according to the reference case scenario for this concept). The container prevents water contacting the wasteform while it remains intact, and provides radiation shielding for the surrounding engineered barrier system materials at early times, when radiation levels are highest, thereby protecting the buffer from radiation damage and radiolysis. Corrosion of the container will also provide redox buffering in the near field, reducing the solubility of some radionuclides following the eventual failure of the container.

**Buffer:** The bentonite buffer swells as it resaturates and thereby provides a low permeability barrier which minimises fluid flow and ensures that solute transport is by diffusion only. Whilst the container remains intact, the bentonite buffer protects it and helps to prolong its life. It does this by limiting the transport of corrosion agents and microbes to the surface of the container, and limiting the transport of corrosion products away from the container. It also provides mechanical support to the container, holding it in position, and protecting it from processes such as rock fall and shear displacements.

Once the container has been breached, the low permeability of the buffer ensures that radionuclide transport from the waste package is diffusion-dominated (slow) and any radionuclide-bearing colloids present are filtered so that their migration is limited. The buffer also provides a medium for radionuclide sorption and helps to buffer the chemical environment to near-neutral to mildly alkaline pH so that radionuclide solubility is minimised.

**Mass backfill:** The disposal tunnels will be completely filled by the containers and bentonite buffer, with no further backfill. The access tunnels will be backfilled with sand mixed with bentonite [144]. This fills void spaces. The backfill also provides a physical barrier to human intrusion.

**Seals:** Multi-component seals in the disposal tunnels can comprise bentonite, rock walls, concrete and gravel; they are designed to resist the swelling pressure of the bentonite buffer, thereby preventing it from expanding and losing density (which would increase its permeability). High integrity seals in access tunnels, composed of concrete or highly compacted bentonite, are designed to prevent these areas from providing preferential pathways for fluid migration.

# 4.2.4 Summary of the evolution of the engineered barrier system for the illustrative concept example for disposal of HLW and spent fuel in a lower strength sedimentary host rock

After resaturation, gradual corrosion of the carbon steel disposal containers will occur. Once they eventually fail, the low permeability rock will provide an important barrier to the migration of radionuclides.

This subsection provides a summary of the key steps in the near-field evolution of the illustrative concept for disposal of HLW and spent fuel in a lower strength sedimentary host rock. The discussion draws in particular on the expected evolution set out by Nagra [33].

The approximate timescales over which key processes dominating the near-field evolution of this illustrative concept are expected to occur are illustrated in Figure 25. Many of the processes identified (such as container degradation, gas generation and buffer alteration) are expected to occur very slowly over many thousands or tens of thousands of years.

These processes can be described independently, but they are strongly coupled to each other.

# Figure 25 Timeframes over which key near-field processes are expected to occur for the illustrative concept example for disposal of HLW and spent fuel in a lower strength sedimentary host rock.



T=0 is closure of the GDF.

### Evolution during construction and operation of the GDF

Construction activities will lead to the formation of an excavation disturbed zone (EDZ) around the tunnels and vaults. The properties of the EDZ will evolve, partly in response to the clay-dominated host rock drying out during the operational phase, and partly because lower strength sedimentary rocks can exhibit creep, particularly if they are poorly indurated. The extent of the EDZ and its characteristics will depend on the geotechnical characteristics of the host rock and the *in situ* stress, both of which are site and depth specific, and on the excavation technique and any excavation support that is installed. In lower strength sedimentary host rocks, such as indurated mudrock, the EDZ generally extends for a distance of up to a tunnel's radius [224]. In the Opalinus Clay, the EDZ is expected to extend about 2 m from the roof and floor of the disposal tunnels [33]. It is expected that damage associated with the EDZ will result in localised changes to hydrogeological properties. Creep, resulting in the self-sealing of fractures and to some degree the excavations themselves, is likely to be important in a lower strength sedimentary host rock [33]. This property is important for performance because it means that the ability of the EDZ to conduct water and contaminants will tend to decrease with time following closure of a disposal vault or tunnel. The timescale for self-sealing will depend on the physical properties of the host rock, the stress regime and the type of engineering support provided. For a poorly-indurated rock, self-sealing and EDZ recovery may be rapid, taking a few years or less to close fractures in the EDZ. However, for more indurated clay rocks, self-sealing could take many centuries or thousands of years, and the disturbed host rock may never completely return to its undisturbed state. The rate and extent of self-sealing is also influenced by the degree to which the rock has become dehydrated.

As water slowly enters the HLW/SF disposal tunnels, the bentonite buffer will swell and seal any gaps between both the waste package and the buffer, and the buffer and the host rock. Prompt sealing of the disposal tunnels is required to ensure that the swelling of the bentonite buffer is resisted so that it achieves its target saturated density, which is required in order to ensure it fulfils its design functions. It should be noted that whilst a limited supply of water in the near field could reduce the rate of bentonite resaturation, it would also limit the corrosion rate of the container by limiting the presence of water and corrosive agents at the container surface.

During this period operational seals<sup>18</sup> may be placed at the entrances to waste-filled disposal tunnels. Given that filling a disposal tunnel will take only a few months, some disposal tunnels will be sealed very early in the operational phase of the GDF. However, the low permeability of the host rock will limit the degree to which the disposal areas are able to resaturate during the operational period. The rate and location of any gas production will need to be taken into account when deciding when and where to emplace seals. Assuming that the excavated cavities will be backfilled progressively during operations, and/or at the time of closure, when the GDF is finally sealed there will be very little remaining void space. The sealing and backfilling materials will have a combination of mechanical properties that will enable them to withstand the stresses caused by creep of the host rock during the operational phase. Seals and backfills using swelling clays such as bentonite will begin to swell, thereby exerting a pressure that will oppose the inward convergence of the host rock. By limiting long-term creep of the host rock, the engineered barrier system prevents macroscopic deformation, including fracturing, and enables a homogeneous stress state to be re-established<sup>19</sup>.

#### Early post-closure evolution

Following closure, the resaturation process described above will continue, and eventually complete. For the Nagra concept modelling work suggests that resaturation may take from ~100 years to many hundreds of years, due to the low hydraulic conductivity of the Opalinus Clay [33].

In addition to the swelling that occurs on resaturation, interaction with the host rock groundwater would result in some minimal changes to the physical and chemical properties of the bentonite buffer. Elevated temperatures will increase the rates of some of these reactions and will influence fluid movements, in particular transport of water vapour, which is an important process in the resaturating bentonite. Porewater redox conditions will tend to be reducing, controlled by the presence of iron in engineered materials, together with a neutral to mildly-alkaline pH provided by the weak buffering capacity of the bentonite and minerals contained in the host rock. In the HLW and spent fuel disposal area, the carbon steel disposal container will provide containment throughout the resaturation period, while the bentonite buffer becomes fully established as a low-permeability barrier, and for a considerable period of time thereafter.

The waste will give off significant quantities of heat until the short-lived component of the inventory has decayed. As a result, the temperature of the buffer and the surrounding rock may rise by several tens of degrees before slowly decreasing. The disposal concept is designed to limit the maximum temperature experienced at the middle of the bentonite barrier to less than ~125°C, to preclude significant thermal alteration that might degrade its desirable swelling and hydraulic properties. Any design temperature limit for the bentonite

<sup>&</sup>lt;sup>18</sup> Temporary seals that could be removed with relatively little effort if required. These could be upgraded or replaced at closure.

<sup>&</sup>lt;sup>19</sup> Such processes may occur during the construction and operational period and throughout the post-closure period.
would thus only apply to this region of the buffer. The maximum temperature in the near field of 140-160°C [33] is likely to occur at the container surface within a few years of waste emplacement and temperatures in the HLW and spent fuel disposal area will remain elevated for over 1000 years. However, it is expected that temperatures should have returned to their undisturbed values before the container stops providing containment. The elevated temperatures in the HLW and spent fuel disposal area may cause a degree of thermally driven mineral redistribution in the near field. The thermal output of ILW may also result in slightly increased (up to about 10°C) temperatures in the ILW disposal area, but this is not expected to significantly affect the evolution of that area.

Despite the relatively benign chemical environment and diffusion-dominated solute transport within the bentonite buffer, corrosion of the carbon steel disposal container will occur [33] and hydrogen gas will be generated. The scientific consensus is that this gas will be able to escape through the bentonite without compromising its properties as a hydraulic barrier, since the bentonite is able to reseal after gas breakthrough [33]. However, since there is little capacity to accommodate gas in the near field of this disposal concept, the gas pressure is expected to accumulate, and eventually will cause two-phase flow of gas (dissolved in groundwater and as a free gas phase) to occur in the near field and the host rock [33].

During this period, the engineered barriers are changing very slowly as a result of interaction with the groundwater and with each other. However they remain sufficiently intact that they are able to perform their intended functions fully. The low-permeability host rock contributes to the durability of the engineered barrier system by providing a stable chemical environment and limiting the rate of water flow through the engineered barrier system. The majority of the degradation reactions that affect the engineered barriers are water mediated, so limiting the volume of water flowing through the near field both limits the rate of barrier degradation and the rate at which dissolved contaminants can be transported away from the disposal containers. Buffers and backfills must be close to full saturation before they can function properly, and slow or uneven resaturation could prevent the barrier functions from becoming established on the expected timescales. Consequently, understanding the movement of groundwater in the near-field and any variability that might be caused by, for example, inhomogeneous rock properties is an important aspect of site investigation.

Radionuclide release from the HLW and spent fuel disposal tunnels while the carbon steel containers remain intact is not expected because the waste packages will continue to provide complete containment. Once the containers fail, the iron corrosion products and the bentonite buffer will provide good substrates for sorption and will retard release from the near field [33]. The low permeability host rock, in which solute transport is predominantly diffusive, will provide an important barrier to the migration of radionuclides that are released from the near field, containing radionuclides so that many decay in the host rock, and do not reach the biosphere.

#### Late post-closure evolution

Based on the current understanding of likely container corrosion rates under *in situ* conditions and material thicknesses, and taking into consideration the chemical and physical protection of the container by the bentonite buffer following resaturation, Nagra expect carbon steel disposal containers to have a lifetime of at least ten thousand years, and possibly longer [33]. Nagra makes a pessimistic assumption that all containers fail simultaneously at 10,000 years after emplacement in their reference case scenario [33].

Once water penetrates the disposal container, the spent fuel and vitrified HLW will start to dissolve and release radionuclides [33]. In the case of the HLW, the majority of the inventory will have decayed by this time; slow dissolution of the glass matrix will limit the release rate of any remaining radionuclide inventory. For the spent fuel, the portion of the

instant-release fraction<sup>20</sup> that has not decayed *in situ* will be released rapidly, and then the spent fuel will slowly dissolve and release the remaining radionuclides [33]. It is expected that the bentonite buffer will provide an effective retardation barrier to the transport of radionuclides from the failed waste package to the host rock.

In the very long term (many hundreds of thousands to millions of years) the disposal container may become penetrated and any radionuclides released will be retarded in the engineered barrier system through sorption onto the bentonite buffer. In a lower strength sedimentary host rock environment, groundwater flow will be diffusion dominated and so the migration of radionuclides away from the near field will be relatively slow. During this time, slow migration of radionuclides out of the engineered barrier system and into the geosphere may occur; hence the engineered barrier system is not expected to provide absolute containment. However, the degraded barriers are likely to continue to retard the release of contaminants to some degree, although by this time the vast majority of the inventory will have decayed.

## 4.2.5 Illustrative concept for disposal of ILW and LLW in a lower strength sedimentary host rock

In this concept, packages of ILW and LLW are placed in reinforced concrete disposal containers, placed in disposal sections separated by concrete bulkheads, within vault-like caverns, backfilled with cementitious mortars.

The Swiss concept for the disposal of ILW and LLW is illustrated in Figure 26. In this concept, packages of ILW/LLW are placed in a reinforced concrete disposal container. Void spaces are filled with cementitious mortar<sup>21</sup>. Some shielded primary waste packages can be directly disposed of, without emplacement in disposal containers. The containers are emplaced in 'disposal sections' separated by concrete bulkheads, within vault-like caverns. These relatively large excavations need to be supported by rock bolts and sprayed concrete linings, together with steel wire mesh reinforcement. Void spaces within the cavern are backfilled with various cementitious mortars as soon as emplacement in a disposal section is complete. Cavern access tunnels are then backfilled with mortar and closed with a concrete plug. Other central access tunnels are backfilled with excavated rock (Opalinus Clay). Various compacted bentonite block seals and concrete plugs are employed to prevent these access ways from becoming preferential pathways for groundwater migration.

<sup>&</sup>lt;sup>20</sup> The portion of the inventory (fission and neutron activation products) that has segregated to the grain boundaries in the wasteform and that may be released rapidly upon exposure to groundwater.

<sup>&</sup>lt;sup>21</sup> This material is different from the NRVB employed in the hard rock example.

## Figure 26 The Nagra concept for the disposal of ILW/LLW in a lower strength sedimentary host rock in Switzerland [144].



Figure 26 illustrates the engineered barriers present in this illustrative disposal concept example.

The primary function of the engineered barriers is to complement the containment provided by the host rock, and to minimise the potential for over-pressurisation in the near field as a result of gas generation due to corrosion. Emphasis is placed on the containment provided by disposal containers and the cementitious mortar (buffer/backfill) [144].

The engineered barriers included in this disposal concept are described below:

**Wasteform:** The wasteform will provide a stable, low solubility matrix that limits the release of the majority of radionuclides by dissolving slowly in groundwaters that come into contact with it [14].

**Container:** The concrete container facilitates waste emplacement and provides a degree of mechanical strength and protection (depending on its thickness). Most are vented, or have a cast cement top surface, to prevent the build-up of excessive gas pressure, and therefore provide only limited physical containment. However, the cementitious mortar used to fill voids around the waste drums within the concrete container has a low hydraulic permeability, thus limiting the rate at which water comes into contact with the waste, and ensuring that radionuclide transport is diffusion dominated, and therefore slow. It also buffers the pH to alkaline values (at which corrosion rates are relatively slow, microbial activity is limited and radionuclide solubility is relatively low) and provides sites for sorption.

**Buffer/backfill**: The cementitious cavern backfill (described in Figure 26 as Mortar 1) provides the same functions as the mortar (described in Figure 26 as Mortar 2) used to fill void space in the disposal containers. It has a low hydraulic permeability, thus limiting the rate at which water comes into contact with the container, and ensuring that radionuclide transport is diffusion dominated, and therefore slow. It also buffers the pH to alkaline

values. The cement-based backfill also has a high capacity for sorbing radionuclides, which further inhibits their migration out of the near field.

The mortar compositions that Nagra plan to employ have a relatively high porosity (up to 50%) [144]. This enables the backfill to accommodate significant quantities of gas generated through corrosion in the near field, thereby minimising the potential for overpressurisation. The backfill also protects the container from rock-fall and provides mechanical resistance to creep closure as it has high compressive strength.

**Mass backfill:** The low permeability of the mortar and excavated rock used to backfill the access tunnels provides a barrier to groundwater flow, preventing the access tunnels from acting as preferential hydraulic pathways, whilst permitting the movement of gases so that over-pressurisation is minimised. The backfill also provides mechanical support to prevent the voids from collapsing as the host rock creeps and the engineered support degrades, and it provides a physical barrier to human intrusion.

**High Integrity Seals:** The concrete plugs and bentonite seals used to close caverns and/or access tunnels are designed to provide a barrier to groundwater flow, preventing these regions from acting as preferential hydraulic pathways, whilst permitting the movement of gases so that over-pressurisation is minimised.

# 4.2.6 Summary of the evolution of the engineered barrier system for the illustrative concept example for disposal of ILW and LLW in a lower strength sedimentary host rock

Following resaturation, high pH conditions will develop that limit solubility and promote precipitation of radionuclides. The low-permeability host rock limits transport of dissolved contaminants.

This subsection provides a summary of the key steps in the near-field evolution of the illustrative concept for disposal of ILW and LLW in a lower strength sedimentary host rock. The discussion draws in particular on the description of the expected evolution set out by Nagra [33, 144].

The approximate timescales over which key processes dominating the near-field evolution of this illustrative concept are expected to occur are illustrated in Figure 27. Many of the processes identified (such as evolution of the backfill mortar) are expected to evolve very slowly over many thousands or tens of thousands of years. These processes can be described independently; however some may be coupled to each other and therefore should be considered within the content of the disposal system.

# Figure 27 Timeframes over which key near-field processes are expected to occur for the illustrative concept example for disposal of ILW/LLW in a lower strength sedimentary host rock.



T=0 is closure of the GDF.

#### Evolution during construction and operation of the GDF

Construction activities will lead to the formation of an excavation disturbed zone around the tunnels and vaults. The properties of the EDZ will evolve, partly in response to the claydominated host rock drying out during the operational phase of the facility lifecycle, and partly because lower strength sedimentary rocks exhibit creep, particularly if they are poorly indurated. The extent of the EDZ and its characteristics will depend on the geotechnical characteristics of the host rock and the *in situ* stress regime, both of which are site and depth specific, and depend on the excavation technique and any excavation support that is installed. In lower strength sedimentary host rocks, such as indurated mudrock, the EDZ may extend for a distance of up to a tunnel's radius [224]. The damage associated with the EDZ is expected to result in localised changes to hydrogeological properties. Creep, resulting in the self-sealing of fractures, and to some extent, the excavations themselves, is likely to be significant in lower strength sedimentary host rock. For a poorly indurated rock, self-sealing and EDZ recovery may be rapid, taking a few years or less to close fractures in the EDZ. However, for more indurated clay rocks, selfsealing could take many centuries or thousands of years, and the disturbed host rock may never completely return to its undisturbed state. The rate and extent of self-sealing are also influenced by the degree to which the rock has become dehydrated.

Backfilling of the disposal vault will increase the temperature (through cement curing) and will bring additional water into contact with the waste packages; some water will have been introduced initially with the raw waste and/or in the cement encapsulant. This is likely to result in a small increase in the rate of corrosion of the waste packages, and hence an increase in the gas generation rate [33, 144], although the rate will be very low because the porewater geochemistry will only favour slow corrosion. As the disposal vault resaturates, any oxygen will be consumed by corrosion and other processes. Alkaline conditions will develop as the incoming groundwater equilibrates with the cementitious backfill.

As soon as they have been sealed the ILW disposal vaults will start to resaturate, albeit slowly, while waste is emplaced in adjacent vaults. Given that filling a disposal vault or

tunnel may take only a few months, some disposal vaults and tunnels will be sealed very early (within the operational phase). However, the low permeability of the host rock will limit the extent to which the disposal areas are able to resaturate during the operational period.

#### Early post-closure evolution

Following closure of the GDF the resaturation process described above will continue, and eventually complete. This is expected to take approximately 500 years, but it could potentially take much longer for the GDF to resaturate fully and for stable, homogeneous conditions to become established, depending on the hydraulic conductivity of the host rock [33]. Gas generated due to corrosion will oppose resaturation by increasing the pressure in the near field. High pH conditions that limit solubility and promote precipitation of key radionuclides will develop in the ILW disposal vaults as the backfill interacts with the incoming groundwater. The wasteform is an important barrier to the release of radionuclides during the resaturation period, while the chemical barrier provided by the cementitious backfill is becoming established.

Significant volumes of gas (mostly hydrogen, but also some methane and carbon dioxide) may be generated in the ILW disposal vaults from the corrosion of the various steel and reactive metal components and the degradation of organic materials in the waste [144]. The gas generation rate may be limited by the availability of water. Carbon dioxide is expected to either dissolve or react with the cementitious backfill. Other gases are likely to accumulate in the near field, within pore spaces in the backfill, initially dissolved in porewater, then as a free gas phase as the concentration increases above the solubility limit. Gas migration through the host rock via diffusion is relatively slow, so it is expected that some pressurisation in the near field will occur as a free gas phase develops. If the gas pressure exceeds the rock entry pressure, free gas could enter the host rock as a result of pore dilation and microfissuring [33, 144]. The Gas status report [5] describes the processes by which free gas may be able to migrate into a lower strength sedimentary host rock once sufficient over-pressures have developed within a GDF. The Nagra concept is designed to ensure that gas pressures cannot build up to the extent that they might damage the integrity of the engineered barriers or the host rock. For example, the high porosity backfill in the ILW vaults is designed to mitigate the localised build-up of excessive gas pressure and suitable seals may allow the passage of gas whilst restricting flow of water.

During this period, the engineered barriers are degrading very slowly as a result of interaction with the host rock groundwater and with each other. However they remain sufficiently intact that they are able to perform their intended functions fully. The low-permeability host rock contributes to the durability of the engineered barrier system by providing a stable chemical environment and limiting the rate of water flow through the engineered barrier system. The majority of the degradation reactions that affect the engineered barriers are water mediated, so limiting the volume of water flowing through the near field both limits the rate of barrier degradation and limits the rate at which dissolved contaminants can be transported away from the waste packages.

During this period, mobile radionuclides as dissolved species will be released from the ILW containers and will generally be retarded within the engineered barrier system.

An alkaline disturbed zone is likely to form around the ILW disposal area, although its extent is likely to be limited. Reactions between cementitious water and the clay minerals of the host rock are expected to result in the precipitation of new minerals that will block the host rock porosity and result in a decrease in permeability and the sealing of any EDZ fractures that have not self-sealed as a result of creep. This will reduce radionuclide transport via the groundwater, but will not have a significant effect on gas migration.

#### Late post-closure evolution

Many tens, or more likely hundreds, of thousands of years after closure, the engineered barriers in the ILW disposal area will begin to lose their effectiveness and will no longer be able to perform their intended functions fully. With time, the wasteforms and the cementitious backfill will alter through reaction with the groundwater and may be disrupted as a result of rock creep and associated compaction. Eventually the pH in the ILW disposal area will fall to a level where the chemical barrier provided by the cementitious backfill loses some of its effectiveness. However, complete degradation of the cementitious barriers is not expected [33]. Those radionuclides that have not decayed within the waste packages and the near field may be released to the host rock. Gas will continue to be produced from the corrosion of metals, but it is expected that the production rate will have significantly declined by this time.

In the very long term (many hundreds of thousands to millions of years) the engineered barrier system is not expected to provide containment. However, the degraded barriers are likely to continue to retard the release of contaminants to some degree, although by this time the vast majority of the inventory will have decayed.

### 4.3 Evolution of the engineered barrier system in a evaporite host rock

Disposal concepts developed in Germany for HLW and spent fuel and in the USA for the disposal of transuranic (TRU) waste are used as illustrative disposal concept examples to demonstrate our understanding of near-field evolution in an evaporite.

Subsection 4.3.1 describes the typical characteristics of an evaporite. The typical requirements on the engineered barrier system are described in subsection 4.3.2.

The concept developed by the US Department of Energy (DOE) for the disposal of transuranic (TRU) waste, is currently being implemented at the Waste Isolation Pilot Plant (WIPP) near Carlsbad, New Mexico. The WIPP is located at 655 m depth in a thick, low-permeability bedded salt (halite) formation that is interbedded with anhydrite and clay. This facility has been operating since 1999 and contains both remote-handled and contact-handled TRU waste packages.

The concept for disposal of HLW and SF in Germany employs crushed rock (salt) as a backfill to provide enhanced physical containment. In contrast, the disposal concept developed and implemented at the WIPP makes use of magnesium oxide (MgO) to provide additional engineered containment by chemically conditioning the near field. In both cases, the engineered barriers are designed to complement the extremely high degree of isolation and containment provided by the host rock.

The key features of these illustrative concepts are described in subsections 4.3.3 and 4.3.5, and the expected evolution of the engineered barriers for these concepts is discussed in subsections 4.3.4 and 4.3.6.

### 4.3.1 Typical characteristics of an evaporite host rock

A key characteristic of evaporites is the ability of the rock to creep and fill voids. There is effectively no groundwater movement. Evaporites typically have a high thermal conductivity.

Two types of evaporite deposits have been considered internationally as host rocks for the disposal of radioactive waste: salt domes and bedded salt deposits. There are many different evaporite minerals, but disposal in rock salt (halite) has received particular attention, partly because of the spatial extent of some halite deposits, and also because of

its favourable mechanical properties, as discussed below. The original concept of geological disposal of radioactive wastes arose from a study carried out by the National Academies in the USA in the 1950s [225], which suggested the use of bedded salt formations. The earliest in situ experimental work on the effects of emplacing heat-emitting wastes into bedded salt formations took place in the USA in the 1960s. Dome salt formations have also been studied extensively in Germany, with extensive design work, full scale engineering testing, safety evaluation and a trial underground facility being constructed at Gorleben, which was intended to be the location of the national GDF for heat-emitting wastes [226]. Dome formations do not however exist in the UK, so our focus is on bedded deposits.

A key characteristic of many evaporites, particularly rock salt, is the ability of the rock to creep at relatively fast rates. If a differential stress exists, for example, if a void space is created in the rock through excavation, an evaporite will deform to fill that void and thereby re-establish isostatic conditions.

As a result of this characteristic, an evaporite host rock has the potential to creep and to seal the openings around and within a GDF and, over time, to provide a complete, uninterrupted barrier around it. This self-sealing behaviour means that any pathways that might enable more rapid migration of fluid, and hence radionuclide transport, away from the engineered barrier system (for example, fractures created in an EDZ) are typically rather short-lived. Consequently, evaporite host rocks are able to provide a high degree of containment over the very long term.

Evaporite deposits typically have the mechanical strength to support relatively large underground excavations (vault-type structures) although rock creep will cause void spaces to close over time. This reduces the range and quantity of materials that will need to be introduced into the GDF for structural support and simplifies the evolution of the near field. However, the creep behaviour of evaporites also affects the extent of waste retrievability that is feasible in this geological environment [75].

In an undisturbed evaporite, although there is water present (for example, about 0.2% by volume at Gorleben [226]) there is effectively no groundwater movement: fractures are largely absent and groundwater cannot flow unless there are significant and connected intercalations of, for example, fractured anhydrite, shale or other porous sediments, within the evaporite sequence. Fluid present within the evaporite units is held within isolated pores or small inclusions, or in intergranular spaces between the evaporite mineral crystals, so there is little accessible water in any one location. Large, isolated brine pockets can occur, but site investigations would aim to identify and avoid these. As a result, any transport of radionuclides will predominantly be diffusive, in intercrystalline fluids, and will occur at very slow rates. Equilibrated groundwater in an evaporite typically has a high salinity and hence is sometimes referred to as 'brine'. Such high-salinity groundwater is due to the relatively high solubility of many evaporite minerals. This means that species may be present in the groundwater at concentrations that might cause enhanced degradation of engineered barriers (particularly by enhancing the rate at which localised corrosion processes occur), compared to that expected in some other geological environments.

Evaporite deposits typically have high thermal conductivity. Consequently, temperature rises in a given location due to heat-emitting waste packages are likely to be lower than in less thermally-conductive rocks. The spacing between heat-emitting waste packages can be closer than in other host rocks because the heat generated is more easily conducted away from the near field and the temperature limit may be higher. This may facilitate a reduced GDF footprint.

### 4.3.2 Typical requirements on the near field in an evaporite host rock

Once rock creep has closed potential pathways for radionuclide migration, an evaporite host rock has the potential to provide a very high degree of containment.

In the short term, prior to self-sealing through rock creep, engineered containment is essential. Once rock creep has closed any potential pathways for enhanced radionuclide migration, an evaporite host rock has the potential to provide a very high degree of containment in its own right. Nevertheless the engineered containment would contribute to a multiple barrier safety concept.

Some form of waste package will be required to facilitate waste emplacement and handling during the operational phase. This could also provide physical containment during the early post-closure phase until creep-closure is complete.

Other engineered barriers could also be employed to provide additional physical and/or chemical containment. Typically, emphasis is placed on engineered seals, which are employed in disposal and/or access tunnels and access shafts. These provide:

- a rigid barrier that imparts structural stability to the surrounding rock, encouraging creep closure. This barrier also reduces the risk of inadvertent human intrusion.
- an impermeable barrier that limits fluid flow and radionuclide transport during the early post-closure period (prior to creep closure). Shaft and tunnel seals may block water-bearing features and ensure that the disposal areas are isolated from surrounding rock formations in which more rapid fluid flow might take place.

In addition, some form of buffer/backfill material (such as crushed host rock in a halite system) may be used to complement the containment provided by the rapid creep-closure of the host rock around the disposal areas by reducing the volume of void space present. Under the influence of rock creep, certain materials used as a buffer or backfill may compact and converge, eventually forming a continuous barrier with the host rock and around the waste packages.

A buffer/backfill might promote the longevity of waste packages by providing some form of physical containment, hence limiting the migration of groundwater and aggressive species to the surface of the waste package. Alternatively (or in addition), a buffer/backfill material could be employed to chemically condition the environment around the waste package and hence, for example could minimise waste package corrosion rates and retard radionuclide migration through the near field.

## 4.3.3 Illustrative concept for disposal of HLW and spent fuel in an evaporite host rock

In this concept, waste in a self-shielded container is emplaced in unlined horizontal tunnels or emplaced in vertical boreholes using thin-walled containers. A crushed salt backfill is used.

Two variant concepts have been developed in Germany (by DBE-Technology GmbH) for the disposal of HLW and spent fuel in an evaporite rock. In the first, waste in a self-shielded container is emplaced in unlined horizontal tunnels excavated in a salt dome host rock. The tunnels are backfilled with crushed salt. The second variant involves the emplacement of HLW and spent fuel in vertical boreholes using thin-walled steel containers. The narrow annulus around the containers is filled with crushed salt. Both disposal concept examples are illustrated in Figure 28.

## Figure 28 Concepts for the disposal of HLW and SF in a salt dome host rock in Germany.



(Left): tunnel-based emplacement concept. (Right): borehole-based emplacement concept. Diagrams courtesy of DBE-Technology GmbH, Germany [91].

Figure 29 illustrates the engineered and natural barriers present in this illustrative concept example, together with their functions. Emphasis is placed on the crushed salt backfill, and on the engineered tunnel and shaft seals.

The engineered barriers included in this disposal concept are described below:

**Wasteform:** In safety assessments for the DBE concept it is assumed that neither the wasteform, nor any fuel cladding present, inhibit radionuclide release. This assumption is conservative, and reflects confidence in the long-term safety provided by other barriers in the geological disposal system. However a wasteform such as vitrified HLW will provide a stable, low solubility matrix that will limit the release of the majority of radionuclides by dissolving slowly, if groundwaters were to come into contact with it [14].

**Container:** The container will provide complete containment during emplacement operations and for a period thereafter.

**Buffer/Backfill:** The crushed salt backfill isolates the container from porewater in the rock. The backfill will compact and creep under the influence of the overburden pressure, as the tunnel walls converge due to the plasticity of salt, and will eventually become continuous, with almost identical properties to those of the surrounding, undisturbed host rock [227]. It therefore encourages rapid sealing of the GDF.

## Figure 29 The multiple barriers present in the illustrative concept example for disposal of HLW and SF in an evaporite host rock.



**Engineered seals:** A series of multi-component shaft and tunnel seals are designed to keep the disposal facility as dry as possible by limiting the quantity of brine available in the near field for radionuclide transport. The seals also ensure that fluid and radionuclides cannot migrate along disposal tunnels and access shafts, (see sections 3.4 and 5.1.2 of [228]). No significant radionuclide releases are expected, even if one of either the tunnel seals or shaft seals fails [228].

Shaft seal designs have been developed in Germany at the Morsleben disposal facility; the designs are composed of bentonite clay, rubble, sand and other materials, and are designed to prevent fluid migration from the surface to the GDF and vice versa, prior to convergence of the host rock The designs also encourage sealing of the facility through creep closure, by providing mechanical stiffness in the shaft column that promotes the early sealing of fractures in the EDZ. The shaft seals also reduce the risk of inadvertent human intrusion.

Tunnel seals have been designed in Germany, incorporating crushed salt, salt briquettes, salt-concrete (concrete made with crushed salt aggregate) and asphalt components. These are designed to prevent fluid entering the tunnels of the GDF (prior to creep closure) [228]. The asphalt provides an impermeable plug over the short to medium term. The salt components encourage sealing through compaction by the surrounding rock, leading to the formation of a continuous salt barrier. One proposed configuration for a tunnel seal developed in Germany is illustrated in Figure 30.

# Figure 30 German tunnel (drift) seal designed for use at the Morsleben disposal facility. Diagram courtesy of DBE-Technology GmbH, Germany [229].



# 4.3.4 Summary of the evolution of the engineered barrier system for the illustrative concept example for disposal of HLW and spent fuel in an evaporite host rock

As the rock creeps, the geological barrier will provide increased containment as time progresses. By the time significant waste package degradation has occurred, complete containment will be provided by the host rock.

This subsection provides a summary of the key steps in the near-field evolution of the illustrative concept example for disposal of HLW and SF in an evaporite host rock. The discussion draws in particular on the description of undisturbed performance included within the 1996 WIPP Compliance Certification Application (see sections 6.0, 6.0.2.2, 6.2 and 6.3.1 of [231], and the subsequent recertification applications [90]). This documentation provides a sound basis for discussion of both the illustrative disposal concept examples for an evaporite host rock.

The behaviour of a disposal system in an evaporite rock is dominated by the coupled processes of deformation of the rock surrounding the excavation, fluid flow, thermal effects and wasteform/waste package degradation. Each of these processes can be described independently, but they are strongly inter-dependent in that the extent to which each process occurs will be affected by the others (see section 6.0.2.2 of [231]).

The approximate timescales over which key processes dominating the near-field evolution of this illustrative disposal concept are expected to occur are illustrated in Figure 32. Many of these near-field processes occur simultaneously and are most pronounced during the early post-closure period.



### Figure 31 Proposed shaft seal design for WIPP [90].

#### Evolution during construction and operation of the GDF

An EDZ will be created as soon as excavation activities commence, as a result of changes in the stress fields in the rock, which lead to brittle fracturing. Rock spalling may also occur during excavation and/or construction. The EDZ will have different mechanical and hydraulic properties to the intact host rock; it will be more extensively fractured, and consequently, will be more permeable, as well as exhibiting a lower pore pressure. It is however likely to extend no more than a few metres from the excavated region.

Resaturation is likely to occur extremely slowly if at all because of the low permeability of evaporites; if diffusive transport dominates, saturation may take many hundreds of years (or longer). Oxidising conditions will predominate in the near field during this phase, but these will quickly revert to reducing conditions once the GDF is closed.

Salt creep will begin immediately following excavation, driven by the presence of differential stress due to the creation of void spaces. Scraping of vault walls, ceilings and floors will be

necessary during construction and emplacement in order to keep facilities open until operations are complete [231]. During construction and operation, any heat generated in the GDF will be dissipated by ventilation and by heat transfer through the surrounding thermally-conductive host rock.

# Figure 32 Timeframes over which key near-field processes are expected to occur for the illustrative concept example for disposal of HLW/SF in an evaporite host rock.



T=0 is closure of the GDF.

#### Early post-closure evolution

After closure, rock creep around the GDF will continue; the geological barrier will provide increased containment as time progresses. Early on, the impermeable components of the shaft seals (such as concrete, clay and asphalt) will be important to prevent inflow of water from the surface to the GDF, as well as to prevent rapid radionuclide transport to the accessible environment. Compaction and creep of crushed salt components of tunnel and shaft seals due to rock creep will render their permeability similar to that of the host rock within approximately 200 years. This will prevent water circulation in the disposal region and to the accessible environment. Rock creep will be accelerated in proximity to heat-generating wastes.

The crushed salt backfill will compact under the influence of surrounding rock creep. Over time, porosity in the crushed salt will be eliminated and eventually the backfill will assume the same properties as the surrounding host rock, merging with it to provide a continuous barrier of low permeability salt.

Relatively rapid recovery of the EDZ is expected through rock creep, which will enable fractures to seal. The recovery of the EDZ around the waste shaft will be aided by the resistance of rigid components of the shaft sealing system. The EDZ around the shaft is therefore not expected to provide a continuous pathway for fluid flow, either from the surface, or out of the near field. Later, the rigid resistance of compacted backfill in the disposal tunnels will also encourage sealing of the fractures surrounding the GDF.

Some waste packages may be breached due to backfill compaction arising from rock creep.

The waste will give off significant quantities of heat until the short-lived component of the radionuclide inventory has decayed. Rock creep is accelerated at higher temperatures. This will encourage more rapid recovery of the EDZ and hence, sealing of the GDF while temperatures are elevated. As a result, complete containment through rock creep is expected after several hundred years for heat-generating wastes, whereas this process is expected to take several thousand years for non-heat-generating waste. Of course, there will also be spatial variations in the creep rate, depending on proximity to heat-generating waste.

As the heat output from the waste falls and the near-field cools, thermal contraction may reduce compressive stresses, or even introduce tensile stresses in the surrounding host rock, which may cause cracking, although this too will heal via creep.

#### Late post-closure evolution

After several thousands of years, the engineered barriers are likely to be significantly degraded. Containers may be breached due to compaction and as a result of various chemical degradation processes. However, by the time significant waste package degradation has occurred and other engineered barriers have also failed, complete containment of the waste in the GDF will be provided by the host rock.

An important requirement for long-term integrity of the GDF is to have ensured, through the site selection and disposal system design processes, that there is no mechanism for preferential transport of fluid (and radionuclides or other possible contaminants) to the surface. If these design and siting requirements are satisfied, then no radionuclide release is expected for a normal (undisturbed) disposal scenario.

## 4.3.5 Illustrative concept for disposal of ILW and LLW in an evaporite host rock

In this concept, shielded transuranic waste is stacked in rectangular-sectioned disposal rooms. Polypropylene sacks of magnesium oxide buffer are placed on top of the stacked waste packages.

In this illustrative disposal concept example, shielded transuranic (TRU) waste is stacked in rectangular disposal rooms, as shown in Figure 33. Remote-handled TRU waste is inserted into horizontal boreholes drilled into the side walls of the disposal rooms before the contact-handled waste is emplaced, as shown in Figure 34. Polypropylene sacks of magnesium oxide buffer are placed on top of the stacked TRU waste packages, as shown in Figure 33. The sacks of magnesium oxide are designed to burst as a result of creep closure and to fill the void space surrounding the waste packages. The sacks are intended to decrease actinide solubilities by consuming any carbon dioxide produced should microbial activity consume the cellulosic, plastic and rubber materials in the waste.

Figure 33 Contact-handled TRU waste packages being stacked in a disposal room in the WIPP with sacks of magnesium oxide backfill placed on top of the waste packages.



Figure 34 Test emplacement of a remote-handled TRU waste container into a horizontal borehole in the wall of a disposal room.



Figure 35 illustrates the engineered and natural barriers present in this illustrative concept example, together with their functions. The engineered barriers included in this illustrative concept are described below:

**Wasteform:** Wastes disposed of at the WIPP are generally not immobilised. However, waste encapsulated in a cement-based wasteform would provide a stable, low solubility matrix that limits the release of the majority of radionuclides by dissolving slowly in groundwaters that come into contact with it [14].

## Figure 35 The multiple barriers present in the illustrative concept example for disposal of ILW and LLW in an evaporite host rock.



**Container:** The container will limit the ability of water to interact with the waste. In addition, steel corrosion products will promote reducing conditions favouring low solubility of actinides. The containers will be vented to prevent gas build-up and over pressurisation [230].

**Buffer/backfill:** Magnesium oxide is a white solid mineral that occurs naturally as periclase. The crystal structure of magnesium oxide is identical to that of sodium chloride (halite), the only credible UK evaporite host rock [15]. It provides the following functions [17]:

- it absorbs carbon dioxide (produced predominantly by degradation of organic matter in the waste) and therefore helps to mitigate the effects of gas generation.
- it is hygroscopic, that is, it absorbs water. This property helps to delay the onset of water contacting waste packages and ultimately wasteforms, thereby delaying radionuclide dissolution and gas generation.
- as it hydrates, magnesium oxide swells, reducing the porosity and permeability of the near field and enhancing the physical containment.
- over time it provides some structural integrity (void filling), through its swelling from any water uptake and once it has been compacted through rock creep.
- it buffers the pH to alkaline values of around pH 9-10. This reduces actinide solubility and container corrosion rates [2, 3] and hence, slows radionuclide migration through the near field.
- it promotes precipitation of actinide-containing minerals as a result of their reduced solubility.
- it sorbs selected radionuclides onto its surface.

**Seals:** Multi-component shaft-seal designs containing concrete, clay, asphalt and salt have been developed for application at the WIPP. They are intended to provide a barrier to fluid flow with a permeability near to that of the undisturbed salt host rock, and to isolate the GDF until rock creep has compacted and consolidated the crushed salt components of the seal. Impermeable elements of the shaft seal, such as clay and asphalt, limit mixing of groundwater between different geological horizons. The proposed shaft seal design for the WIPP is illustrated in Figure 31. Over time, the crushed salt components will compact and

develop properties similar to those of the host rock, ensuring that an uninterrupted barrier of low permeability salt is present around the entire GDF [90].

Panel (tunnel) seals have also been designed for application in the WIPP. However, these are intended to address operational safety requirements; they are not designed to contribute to long-term (post-closure) safety [231].

# 4.3.6 Summary of the evolution of the engineered barrier system for the illustrative concept example for disposal of LLW and ILW in an evaporite host rock

As the rock creeps, the geological barrier will provide increased containment as time progresses. By the time significant waste package degradation has occurred, complete containment will be provided by the host rock.

This subsection provides a summary of the key steps in the near-field evolution of the illustrative concept example for disposal of ILW and LLW in an evaporite host rock. As for the previous section, the discussion draws in particular on the description of undisturbed performance included within the 1996 WIPP Compliance Certification Application (CCA) (see sections 6.0, 6.0.2.2, 6.2 and 6.3.1 of [231], and subsequent recertification applications [90]).

The behaviour of a disposal system in an evaporite is dominated by the coupled processes of deformation of the rock surrounding the excavation, any fluid flow, thermal effects and wasteform/waste package degradation. Each of these processes can be described independently, but they are strongly inter-dependent; the extent to which each process occurs will be affected by the others (see sections 6.0.2.2 of [231]).

The approximate timescales over which key processes dominating the near-field evolution of this illustrative disposal concept are expected to occur are illustrated in Figure 36. Many of these near-field processes occur simultaneously and are most pronounced during the early post-closure period.

# Figure 36 Timeframes over which key near-field processes are expected to occur for the illustrative ILW/LLW disposal concept in an evaporite host rock.



T=0 is closure of the GDF.

#### Evolution during construction and operation of the GDF

The evolution of the EDZ in an evaporite has been previously described in subsection 4.3.4.

Saturation is likely to occur extremely slowly, if at all, because of the low permeability of salt; if diffusive transport dominates, resaturation may take many hundreds of years (or longer).

Salt creep will begin immediately following excavation, driven by the presence of differential stresses due to the creation of void spaces. Scraping of vault walls, ceilings and floors will be necessary during construction and emplacement in order to keep facilities open until operations are complete. During construction and operation, any heat generated in the GDF will be dissipated by ventilation and by heat transfer through the surrounding host rock. Oxidising conditions will predominate in the near field during this phase, but these will quickly revert to reducing conditions once the GDF is closed.

The magnesium oxide buffer will start to absorb moisture and carbon dioxide as soon as it is emplaced, and in the longer-term will buffer the pH of the near field after closure. There will be sufficient magnesium oxide present to consume all the carbon dioxide that would be produced if all the organic materials in the waste were to be consumed by microbial activity. In the WIPP, the functionality of the magnesium oxide buffer processes are initially limited by the polypropylene 'supersacks' used during emplacement. Ventilation of the GDF during construction and operation will help to maintain the function of the buffer prior to sealing of the vault by removing some of the moisture and carbon dioxide from the GDF.

#### Early post-closure evolution

Rock creep around the GDF will continue; the geological barrier providing increased containment as time progresses. Early on, the impermeable components of the shaft seals (such as concrete, clay and asphalt) will be important to prevent inflow of water from the surface to the GDF, as well as to prevent rapid radionuclide transport to the accessible

environment. Compaction and creep of crushed salt components of tunnel and shaft seals due to rock creep will render their permeability similar to that of the host rock within approximately 200 years. This will prevent water circulation in the disposal region and to the accessible environment. Cooling of the host rock in the vicinity of cementitious seal components, following heat output due to cement curing, may cause localised cracking of the evaporite due to thermal contraction. However, this is unlikely to be spatially extensive and, over time, will recover through rock creep.

Relatively rapid recovery of the EDZ is expected through rock creep, which will enable fractures to seal. The recovery of the EDZ around the disposal shaft will be aided by the resistance of rigid components of the shaft sealing system. The EDZ around the shaft is therefore not expected to provide a continuous pathway for fluid flow. Later, the rigid resistance of compacted backfill in the disposal tunnels will also encourage sealing of the fractures surrounding the GDF.

Rock creep and the collapse of excavations will cause the magnesium oxide sacks to burst and spread as a powder around the waste packages. The disposal areas will be gradually compacted as further salt creep occurs. The magnesium oxide will continue to fulfil its chemical buffering functions through the early post-closure phase, minimising free water in the near field and protecting the waste package from chemical degradation.

Some waste packages may be breached due to the convergence of the walls, floors and ceiling of excavated rooms, together with compaction of the magnesium oxide buffer arising from host rock creep.

#### Late post-closure evolution

After several thousands of years, the engineered barriers are likely to be significantly degraded. Multiple breached waste packages are expected due to compaction and as a result of various chemical degradation processes. It is unlikely that the magnesium oxide will provide an effective buffering capability to reduce radionuclide solubility and to promote package longevity over the very long term (hundreds of thousands of years). However, by the time significant waste package degradation has occurred and other engineered barriers have also failed, complete containment of the waste in the GDF will be provided by the host rock. An important requirement for long-term integrity of the GDF at this point is to have ensured, through the site selection and disposal system design processes, that there is no mechanism for preferential transport of fluid (and radionuclides or other possible contaminants) to the surface. If these design and siting requirements are satisfied, then no radionuclide release is expected for a normal (undisturbed) disposal scenario.

### 5 Concluding remarks

The science and technology underpinning geological disposal of the materials currently considered in the UK radioactive waste inventory is well established. The knowledge base includes information from laboratory studies, demonstration experiments, models and studies from archaeological and natural analogues that can be used to support the implementation of geological disposal.

The key message emerging from the analysis presented in this status report is that it is important to understand which key processes affecting evolution of the engineered barrier system are likely to occur, how important these processes are for different disposal concepts and different geological environments, and the order and timescales over which they occur. Specifically:

- Processes that occur during the construction and operational period that will need to be considered when developing site-specific understanding of the evolution of the engineered barriers include creation and partial or complete recovery of an excavation disturbed zone and desaturation and oxidation of part of the host rock immediately surrounding the GDF.
- Post-closure processes that may contribute to the evolution of the engineered barrier system once wastes and the buffer and/or backfill materials have been emplaced and disposal areas sealed include heat generation and thermal effects, irradiation of engineered barrier system materials, host rock creep, degradation of buffer and backfill materials, gas generation, microbial effects and waste container evolution.
- The specific processes occurring within the evolving engineered barrier system of a GDF will depend upon a number of factors, including the nature of the waste, the choice of engineered materials used in components of the near field, and how these interact with each other, with the waste and with the surrounding host rock, and the nature of the geological environment and groundwater.
- The engineered barrier system provides long-term containment of radionuclides, thereby limiting their release to the geosphere. Over time, the barriers will evolve as they interact with each other and with their surroundings. They will however still continue to fulfil their functions for many thousands to hundreds of thousands of years.

Information contained in the suite of Research Status Reports has been used to underpin the development of the 2016 generic DSSC. In particular, information from this status report has been used to provide technical underpinning to the expected evolution of the engineered barrier system before and after emplacement in the GDF.

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