THE USE OF CLAY AS AN ENGINEERED BARRIER IN RADIOACTIVE-WASTE MANAGEMENT – A REVIEW

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Abstract—Geological disposal is the preferred option for the final storage of high-level nuclear waste and spent nuclear fuel in most countries. The selected host rock may be different in individual national programs for radioactive-waste management and the engineered barrier systems that protect and isolate the waste may also differ, but almost all programs are considering an engineered barrier. Clay is used as a buffer that surrounds and protects the individual waste packages and/or as tunnel seal that seals off the disposal galleries from the shafts leading to the surface.

Bentonite and bentonite/sand mixtures are selected primarily because of their low hydraulic permeability in a saturated state. This ensures that diffusion will be the dominant transport mechanism in the barrier. Another key advantage is the swelling pressure, which ensures a self-sealing ability and closes gaps in the installed barrier and the excavation-damaged zone around the emplacement tunnels. Bentonite is a natural geological material that has been stable over timescales of millions of years and this is important as the barriers need to retain their properties for up to 10^6 y.

In order to be able to license a final repository for high-level radioactive waste, a solid understanding of how the barriers evolve with time is needed. This understanding is based on scientific knowledge about the processes and boundary conditions acting on the barriers in the repository. These are often divided into thermal, hydraulic, mechanical, and (bio)chemical processes. Examples of areas that need to be evaluated are the evolution of temperature in the repository during the early stage due to the decay heat in the waste, re-saturation of the bentonite blocks installed, build-up of swelling pressure on the containers and the surrounding rock, and degradation of the montmorillonite component in the bentonite. Another important area of development is the engineering aspects: how can the barriers be manufactured, subjected to quality control, and installed?

Geological disposal programs for radioactive waste have generated a large body of information on the safety-relevant properties of clays used as engineered barriers. The major relevant findings of the past 35 y are reviewed here.

Key Words—Bentonite, Barrier, Nuclear Waste, Geological Disposal, Waste Management.

INTRODUCTION

Worldwide, no final repository for high-level radioactive waste has been constructed as yet, though many countries have had extensive research programs running since the 1970s to develop safe and sustainable repository concepts for high-level waste. International consensus (IAEA, 2003) has proposed geological disposal as the safest option and this method will, therefore, be the focus of this review.

Geological disposal was defined in a 1995 Collective Opinion of the Nuclear Energy Agency (NEA) Radioactive Waste Management Committee entitled 'The Environmental and Ethical Basis of Geological Disposal' (OECD NEA, 1995). According to page 16 of that publication, geological disposal is provided by a system that will: (1) "isolate the wastes from the biosphere for extremely long periods of time"; and

(2) "ensure that residual radioactive substances reaching the biosphere will be at concentrations that are insignificant compared, for example, with the natural background levels of radioactivity." Geological disposal should also "provide reasonable assurance that any risk from inadvertent human intrusion would be very small."

The isolation of the waste from the biosphere relies, in most repository concepts, on passive multi-barrier systems. These typically comprise the natural geological barrier provided by the repository host rock and its surroundings and an engineered barrier system (EBS). This multi-barrier principle creates an overall robustness of the system that enhances safety by containing the waste in different materials with different properties, *e.g.* corrosion-resistant materials that isolate the waste or low-permeability materials that restrict groundwater movement. As part of the EBS concept, bentonite or bentonite-sand mixtures are often selected as a buffer between the waste canister and the host rock or as seals in excavated disposal galleries. An overview is given here of why and how bentonite is used as a buffer in different repository concepts. This will cover:

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(1) The favorable properties of bentonite for nuclear-waste disposal. To what extent does bentonite contribute to safety and what safety functions does bentonite have to fulfill?

(2) The use of bentonite in different national concepts.

(3) How does bentonite perform over very long timescales? What are the scientific bases? What are the challenges and uncertainties?

(4) Engineering aspects: how is the bentonite barrier manufactured and installed?

Examples will be given from the Swedish program, where bentonite is used in a crystalline host rock, and the Swiss program, where bentonite is used in a clay host rock.

DESIRED PROPERTIES – SAFETY FUNCTIONS

To contain the waste safely and comply with the overriding safety principles, the key requirements of a buffer material in the case of high-level nuclear waste disposal, independent of host rock, are: (1) a low hydraulic permeability/conductivity; (2) a self-sealing ability; and (3) durability of properties over the very long term. The second requirement rules out brittle materials like concrete, while the third rules out organic materials like asphalt. Swelling clay, therefore, stands out as a viable buffer material and bentonite has, thus, been selected as a barrier material in basically all national programs that are looking at the option of disposing of high-level waste in fractured rock below the groundwater table. One example of this is the Swedish KBS-3 concept (Figure 1). The overall safety functions

of a nuclear-waste repository are generally confinement and retardation. Confinement is defined as the complete isolation of the waste, while retardation slows down any releases in the case of failed confinement.

The overall criterion for evaluating repository safety is issued by the national regulator and is usually expressed as a maximum dose, or risk, to a representative individual in the group exposed to the greatest risk. In order to evaluate the dose or risk from a repository, a detailed and quantitative understanding of all processes that affect the repository, together with the associated uncertainties, is needed. Dose and risk are, therefore, not very practical factors to use for the study of individual repository components. To resolve this, the concept of 'safety functions' has been introduced. A safety function is a description of how an individual barrier contributes to confinement and retardation. Safety functions can be defined based on the understanding of the properties of the components and the long-term evolution of the system.

LIMITING ADVECTIVE TRANSPORT IN THE NEAR-FIELD

An important safety function of the buffer is to limit the transport of dissolved species that could cause canister corrosion and potential radionuclide release from a damaged canister. The material of the buffer surrounding the canister is selected so as to prevent advective transport in the near-field. A guideline is that the hydraulic conductivity of the saturated buffer should be sufficiently low to ensure that diffusion is the dominant transport mechanism. Depending on the
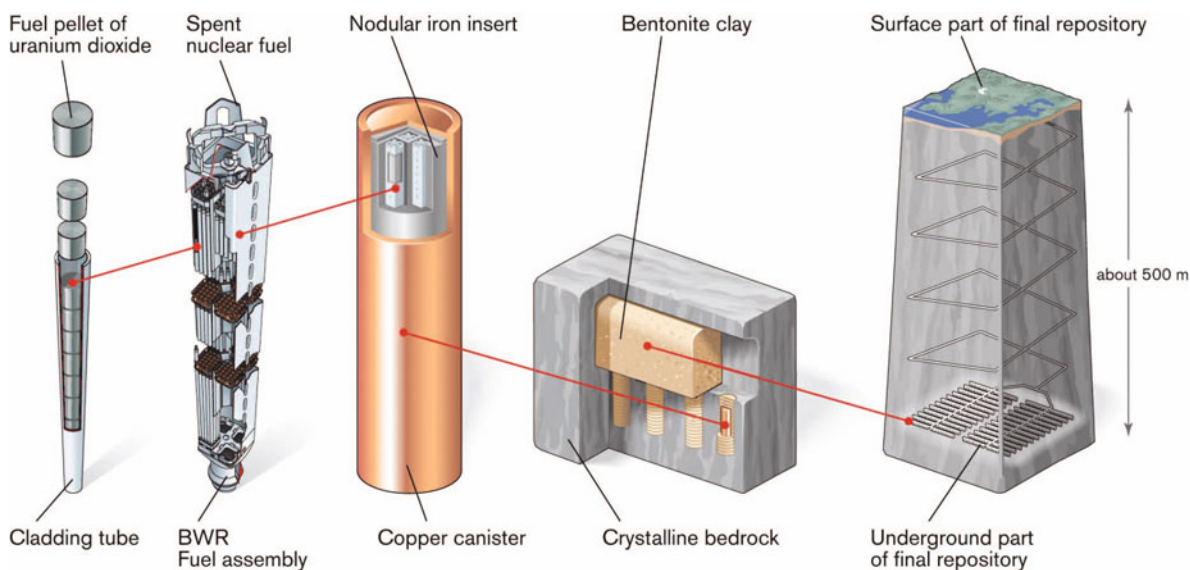

Figure 1. The KBS-3 method. The method involves encapsulating the spent fuel in copper canisters which are then emplaced, surrounded by a buffer of bentonite clay, in deposition holes in a tunnel system at a depth of 400−700 m in the bedrock (SKB, 2011, reproduced with permission). BWR: boiling water reactor.

concept and the boundary conditions, a hydraulic conductivity of $10^{-11}-10^{-12}$ m s^{-1} is required to fulfil the safety function.

The buffer homogeneity is ensured partially by the fact that the buffer consists of a clay material that swells when saturated with water. The swelling enables the installed clay to fill engineering gaps and voids as well as to balance the buffer mass between components installed with different initial densities (e.g. compacted bentonite blocks and bentonite pellets). A swelling pressure criterion is, therefore, formulated. The safety function is usually considered to be upheld if the swelling pressure is in the range of 0.1−1.0 MPa. The swelling pressure also ensures a self-sealing ability of the buffer.

REDUCING MICROBIAL ACTIVITY

Sulfide production by sulfate-reducing bacteria initially present in the buffer and in the host rock or introduced during the realization of the near-field is, in the long term, normally restricted to insignificant levels by the reliance of the bacteria on nutrients present in the groundwater. In certain transient situations, the access to nutrients could be significant, e.g. due to degradation of construction and stray materials in the repository, cement additives (such as superplasticizers), organic components present in the pore water of sedimentary host rock, or even hydrogen gas produced by the anaerobic corrosion of metal components. In such cases, the buffer has the function of reducing the activity of initially present or introduced microbes.

The microbial activity can be limited by increasing swelling pressure, which can be achieved by increasing material density and, thus, reducing the pore size and diffusivity of nutrients. The quantitative treatment of a situation of this type would, however, depend on a number of factors, meaning that while the buffer density or swelling pressure are useful indicators for this buffer function, a strict criterion for buffer density cannot be formulated.

DAMPING ROCK-SHEAR MOVEMENTS

Another safety function of the buffer is to protect the canister from rock movements, especially from the consequences of rock-shear movements caused by earthquakes. The plasticity of the bentonite enables the buffer to mitigate shear significantly. This is of greatest relevance for a crystalline host rock and depends on a number of factors, mainly the properties of the canister materials and canister design, the buffer thickness, and the shear movements anticipated at the given site. An increased buffer density and swelling pressure reduces the plasticity. In the Swedish case, for a maximum shear movement of 50 mm a design premise of a maximum installed density of 2050 kg/m^3 (for a saturated buffer) has been established. A greater density would result in a

significant increase in the shear forces on the canister in the case of rock movements.

RESISTING TRANSFORMATIONS (TEMPERATURE REQUIREMENT)

To avoid transformation of the montmorillonite component in the buffer to non-expandable minerals, a temperature safety function can be defined. However, as the heat generation from the waste decreases relatively rapidly with time, and montmorillonite alteration is a slow process, any temperature criteria will always be somewhat arbitrary. Still, for repositories in crystalline rock (e.g. Sweden, Finland) a temperature limit of 100ºC is set in order to limit chemical alterations. In clay environments higher temperatures can be tolerated as the host rock itself is the primary barrier.

PREVENTING CANISTER SINKING

The swelling pressure of bentonite should be sufficiently high that it prevents the canister from sinking through the supporting bentonite layers to avoid any direct contact with the host rock (or the concrete bottom plate in a deposition hole) and short-circuiting of the buffer.

The main determinant of the creep rate and the resulting canister sinking is the magnitude of the mobilized shear strength (shear stress divided by shear strength), which results in increased canister sinking (Dueck et al., 2010). The shear strength decreases with decreasing swelling pressure. Analyses of canister sinking in a deposition hole for a range of buffer densities, and hence swelling pressures, indicate that the total sinking will be <2 cm for swelling pressures down to 0.1 MPa.

LIMITING PRESSURE ON CANISTER AND ROCK

The maximum swelling pressure from the bentonite buffer must be limited in order to avoid mechanical damage to the canister and the surrounding host rock. The pressure permitted depends on the properties of the barrier. Under the initial conditions, the buffer is installed with a known density and moisture content and is unsaturated. Upon saturation by inflowing groundwater, it swells and reaches a maximum swelling pressure. This places strict requirements on the manufacturing and installation process, as the swelling pressure is exponentially dependent on the density (e.g. Wang et al., 2012; Karnland et al., 2008; Dixon et al., 1996). A small deviation in the mass of the installed buffer may consequently lead to a large increase in swelling pressure.

In the Swedish case, the design premise of the isostatic load on the canister was determined under the assumption that the buffer swelling pressure will not exceed 15 MPa. If the buffer freezes, development of

damaging pressures due to expanding water cannot be ruled out. Therefore, the buffer temperature should not fall below the freezing temperature of a water-saturated buffer. This process is relevant only for repositories in areas where severe permafrost is expected in the future. The freezing point of bentonite is related to the swelling pressure (discussed later in this document). The swelling pressure of the buffer should, therefore, be sufficient to ensure that freezing never occurs. A swelling pressure of 5 MPa would yield a freezing point of $\sim-4°C$, which is below the lowest temperature expected at the selected repository sites in Sweden and Finland, even for a very pessimistic assumption about future climate evolution.

In a sedimentary host rock, the upper limit for the swelling pressure is given by the effective stress of the overburden. By exceeding the effective stress of the overburden, the sedimentary bedding structure could be damaged.

OTHER REQUIREMENTS

The amount of canister-corroding agents in the buffer should be small. Apart from unavoidable initial amounts of oxygen, the pyrite content could also support corrosion because, if not oxidized by oxygen initially present or intruding, it will release sulfide, a canister-corroding agent. If microbial reduction of sulfate cannot be avoided, sulfate-containing minerals like gypsum and anhydrite could also pose a problem. Most of the other accessory minerals in the bentonites, quartz, feldspars, other clay minerals, *etc.*, are not expected to have any detrimental effects on the repository.

SUMMARY

Based on these requirements, the selected reference buffer material is a bentonite clay which fulfills two basic mineralogical criteria: firstly, the montmorillonite content must be sufficiently high to uphold and maintain the minimum swelling pressure, to ensure sufficiently low hydraulic conductivity, and to ensure the necessary range for stiffness and shear strength for the specified density interval; secondly, the amount of detrimental accessory minerals must be small.

The selected reference bentonite qualities and densities vary slightly for different national concepts, but the dry density is generally in the range of $\sim1450-1650$ kg/m^3 for a fully saturated and homogenized buffer in the deposition hole or tunnel.

NATIONAL CONCEPTS – A BRIEF OVERVIEW

Bentonite is an integral part of the engineered barrier system in many national concepts for high-level nuclear-waste disposal. The following section gives a brief overview and some examples of the engineered barriers in several national programs.

Sweden and Finland

The Swedish concept for geological disposal was developed in the mid-1970s. The original concept was to dispose of vitrified high-level waste in titanium containers surrounded by a 10/90 bentonite/sand mixture as a buffer at a depth of ~500 m in the bedrock. In 1978, the direct disposal of spent fuel was investigated and the titanium container was changed to copper and the bentonite/sand buffer was replaced with pure bentonite. Copper was introduced to obtain a very long service lifetime in an oxygen-free environment.

At this early stage, Wyoming-type natural Na-bentonite was selected as the reference material. The buffer was assumed to be installed as isostatically compacted blocks surrounded by bentonite powder to fill any voids in the system.

This concept has remained virtually unchanged, but is now called KBS-3 (Figure 1) and was used in the applications submitted for a permit to build a final repository for Sweden's spent nuclear fuel in Forsmark (SKB, 2011).

Preparations for the final disposal of spent nuclear fuel in Finland began at the same time as the commissioning of the first nuclear power plants in the late 1970s. The schedule for final disposal was set in 1983, when the Finnish government decided on the objectives and program for nuclear-waste management. In 1994, the Nuclear Energy Act came into force, according to which all nuclear waste must be treated, stored, and disposed of in Finland, and no nuclear waste from other countries would be imported into Finland. Next, Imatran Voima and Teollisuuden Voima established Posiva Oy to implement the final disposal of spent nuclear fuel and carry out the associated research. An application for a decision-in-principle from the government was submitted by Posiva in 1999. In this application, Posiva proposed Olkiluoto in Eurajoki as the site for the repository and KBS-3 (Figure 1) as the chosen method for implementing final disposal.

Canada

Canada has focused its research and development efforts for the long-term management of high-level nuclear waste on the concept of Deep Geological Disposal. In 1975, the Canadian nuclear industry defined its waste management objective as being to "...isolate and contain the radioactive material so that no long-term surveillance by future generations will be required and that there will be negligible risk to man and his environment at any time .... Storage underground, in deep impermeable strata, will be developed to provide ultimate isolation from the environment with the minimum of surveillance and maintenance."

In 1978, the governments of Canada and Ontario jointly established the Canadian Nuclear Fuel Waste Management Program (CNFWMP). Under the program,

the federal government, through its crown corporation, Atomic Energy of Canada Ltd. (AECL), had responsibility for managing the program and developing the technology for long-term disposal of used nuclear fuel. In 1988, the CNFWMP, through AECL, submitted its generic (non-site-specific) proposal (AECL, 1994) for long-term management of used nuclear fuel. Under the proposal, the used fuel would be placed in disposal vaults ~500−1000 m deep in the granite rock of the Canadian Shield. The fuel would be encased in corrosion-resistant containers designed to last thousands of years and surrounded by a buffer material (such as bentonite clay) that retards water migration. The vaults, tunnels, and shafts of the disposal site would be backfilled and sealed during the closure stage.

Today, Canada's long-term used nuclear fuel-management program is administered by the Nuclear Waste Management Organization (NWMO). Canada's plan for the long-term care of used nuclear fuel is known as 'Adaptive Phased Management.' Used fuel will be contained and isolated in a deep geological repository in a suitable rock formation using a multiple barrier system (Figure 2). Site selection is based on screening of potential sites of volunteer communities followed by a preliminary assessment of potential suitability.

Spain

Since 1987, the National Spanish radioactive waste management organisation, ENRESA (Empresa Nacional de Residuos Radiactivos), has been developing a disposal program aimed at providing a final solution for spent fuel and high-level waste. The program comprises three major areas of activity: identification of suitable sites, conceptual design, and performance assessment of a geological repository, as well as research and development.
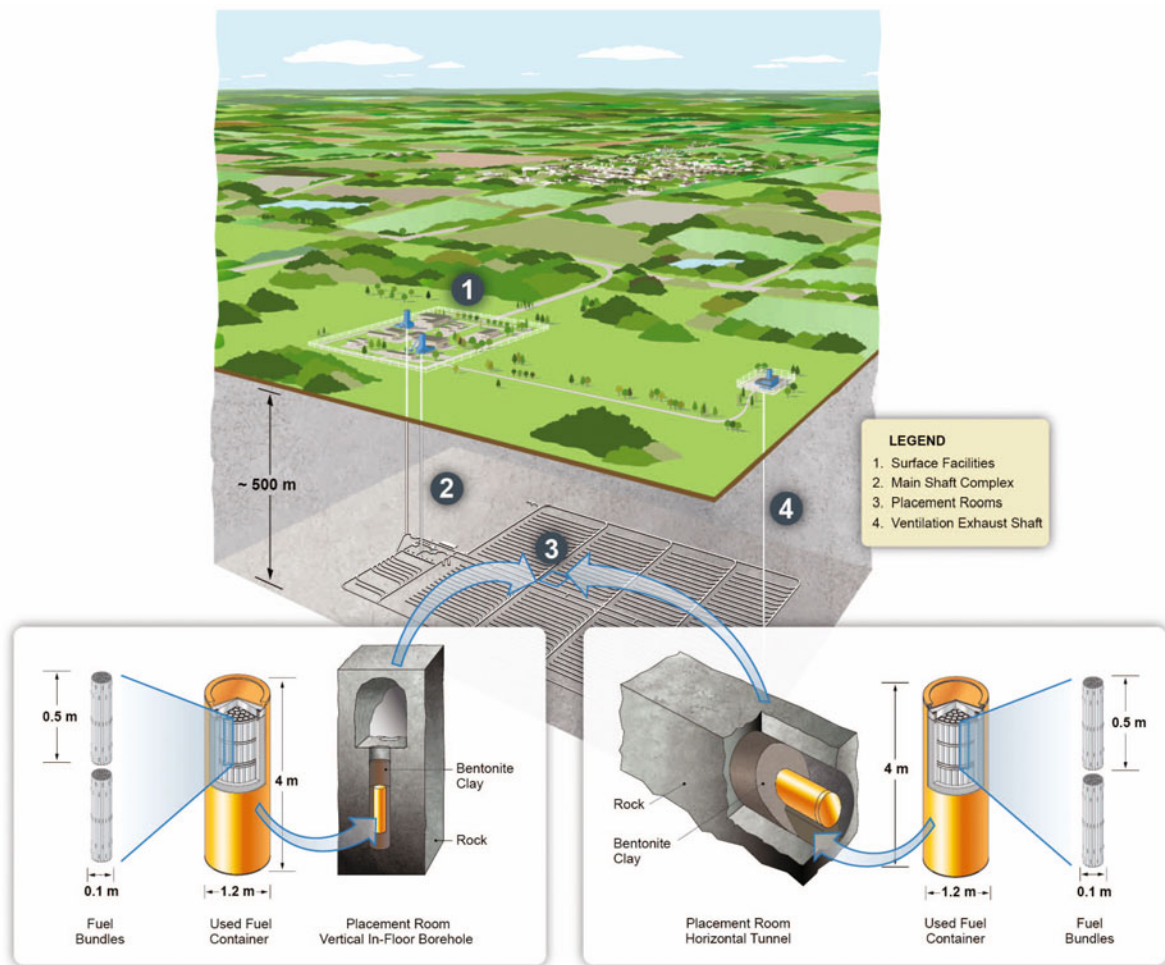

Figure 2. Two options for disposal according to the Canadian concept: the containers will be placed in vertical boreholes drilled in the ground along the axis of a placement room, or placed horizontally within the confines of a placement room, depending on the nature of the rock and characteristics of the site (from Nuclear Waste Management Organization, 2012; reproduced with permission).

Non-site-specific conceptual repository designs have been developed for three candidate host rocks (clay, granite, and salt) in order to provide a basis for the research and development activities and the performance and safety assessment studies of the repository system (Figure 3).

Japan

In Japan, the focus of strategies for high-level waste management has been on geological disposal. The current safety concept places most weight on the near-field (the engineered barrier system and a restricted zone of the surrounding geosphere) (Figure 4). When a potential disposal site is selected in the future, an optimized design for the engineered barrier system will be determined in light of safety and economic considerations, which take into account the geological conditions specific to the site (JNC, 2000).

Switzerland

NAGRA is responsible in Switzerland for the safe disposal of all radioactive waste. Demonstration of the disposal feasibility for high-level waste was achieved with the project 'Entsorgungsnachweis' (NAGRA, 2002). Within the framework of the Sectoral Plan for Deep Geological Repositories, under the lead of the Federal Office of Energy, NAGRA screened Switzerland's geological underground for adequate geological situations, potential host-rock options, and siting regions (NAGRA, 2008) and submitted the results to the authorities in 2008. After an extensive review process, all six geological siting regions proposed by NAGRA were accepted by the Swiss government in 2011. Stage 2 of the site selection process, now on-going, will result in the proposal of at least two sites each for the low- and intermediate-level waste and high-level waste repositories.

The concept foreseeing the use of bentonite in the near-field as a backfill and buffer material in the horizontal emplacement concept for HLW was described in 'Entsorgungsnachweis' (NAGRA, 2002). Bentonite was selected as the buffer and backfill material on the basis of early scientific work carried out at the Institute for Geotechnical Engineering at ETH Zürich, high-lighting the favorable mechanical and hydrological properties of swelling clay (*e.g.* Madsen and Müller Vonmoos, 1989; Madsen, 1998).

The repository concept in Opalinus Clay proposed by NAGRA (2002) as part of the 'Entsorgungsnachweis' project was adapted to use pellets around the canisters, which are supported by bentonite blocks as proposed in earlier studies.

NAGRA's repository concept in the Opalinus Clay envisions an array of long (up to 800 m) parallel tunnels at a depth of 600−900 m containing SF or HLW canisters, with the region around the canisters filled with bentonite (Figure 5). The canisters would be constructed from thick (12−14 cm) carbon steel (Carbon steel is steel in which the main interstitial alloying constituent is carbon in the range of 0.12−2.0%. The term 'carbon steel' is often used in reference to steel which is not stainless steel; in this use carbon steel may include alloy steels.) or possibly with a copper shell. The bentonite buffer has a reference saturated density of ~1.9 mg m^{-3}. The Opalinus Clay has a low hydraulic conductivity in the range of 10^{-12}−10^{-14} m s^{-1} and a porosity of 0.12−0.15. The Opalinus Clay formation is generally >100 m thick and is surrounded by clay-rich confining units. The repository is connected with the surface *via* shaft and/or ramp; these are also backfilled partially with sand/bentonite mixtures.

A low-pH shotcrete liner or other tunnel-supporting measures are foreseen, with a hydraulic seal placed between every tenth canister. The hydraulic seal would comprise bentonite of greater density than that used around the disposal canisters, constructed using a combination of blocks and pellets. In the seal zone,
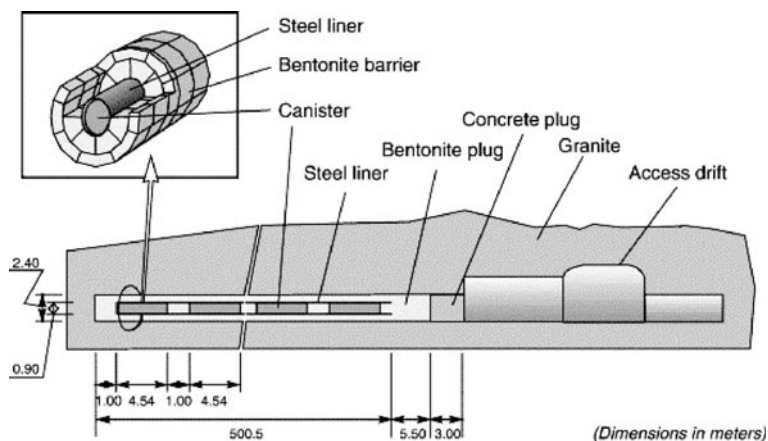

Figure 3. Longitudinal section of a disposal drift for the Spanish reference granite case (from ENRESA, 2000; reproduced with permission).

H12 disposal concept

Vitrified Waste
(in stainless steel canister)

- Uniform and stable confinement of radionuclides in glass matrix
- Mitigation of radionuclides to groundwater due to chemical durability
- Stable for thermal and radiation impact

Overpack
(Carbon steel)

- Physical containment of vitrified waste during the period of high heat generation and radiation
- Ensures reducing conditions around the vitrified waste
- Sorption of radionuclides onto corrosion products

Buffer
(Mixture of bentonite and sand)

- Low permeability
- Low mass transport
- Retardation of radionuclide migration (Sorption)
- Swelling and plasticity
- Chemical buffering
- Low solubility in porewater
- Filtration for colloid migration, organic matter, and microbes

Geological Environment
Long-term stability

- No volcanic activity
- No active fault
- No significant effect due to uplift/denudation
- No significant effect due to climate/sea-level change
- Presence of natural resources

Favorable geological environment

- Favorable geochemistry (*e.g.* chemically reducing)
- Low groundwater flux
- Rock mechanical stability
- Isolation from the human environment

Function as a natural barrier system

- Retardation and dilution/dispersion for radionuclide migration
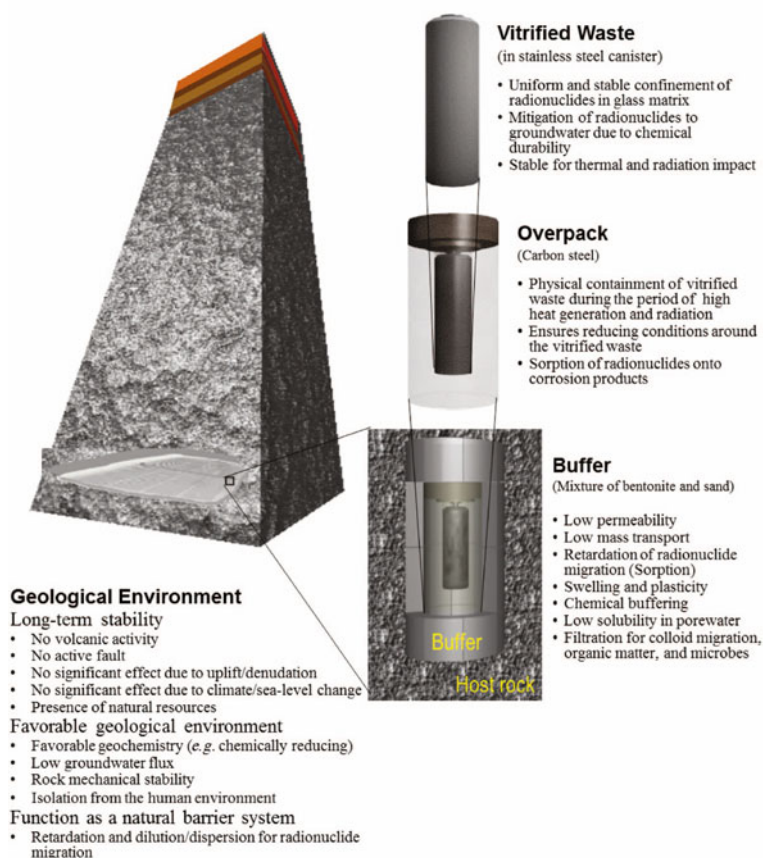
Figure 4. Basic concept of the geological disposal system in Japan and its expected barrier functions (from JNC, 2000; reproduced with permission).

steel ribs would be used for support rather than a shotcrete liner. The purpose of eliminating the liner in the hydraulic seal zone is to prevent any possibility of hydraulic flow through the more permeable liner (NAGRA, 2010).

A mixture of sand and bentonite is foreseen as a sealing material for the emplacement tunnels and caverns at different locations in the repository. Favorable and safety-relevant properties of a sand-bentonite mixture are the relatively high gas permeability and intrinsic low hydraulic conductivity of this material.

France

For the disposal of HLW, ANDRA (National Agency for the Management of Radioactive Waste) is working towards reversible disposal in a deep geological repository ('CIGEO' project), planned to commence in 2025. The disposal facilities are to be developed on the basis of the 500 m-deep research laboratory (LSMHM Underground Research Laboratory) in the Callovo-Oxfordian clay formation at Bure in the Meuse district in eastern France.

This facility was licensed on August 3, 1999, and its construction (access shafts, basic drift network with underground ventilation) was completed in 2006. Nevertheless, more drifts and niches are due to be excavated for the on-going geological survey and experimental program or the engineering-technology demonstrations. Apart from this underground research laboratory, ANDRA has commissioned a Technological Exhibition Facility (ETe), in order to design and operate prototypes and demonstrators. The ETe has been open to the public since mid-2008.

After a dialogue phase with local stakeholders and approval by the government in 2010, ANDRA has also focused, within the 250 km² so-called 'transposition zone' initially defined, on a 30 km² area for more detailed investigations to locate the underground facilities of the CIGEO repository. A similar process, within the framework of a public debate, should specify the location of the various surface facilities.

In 2012, ANDRA awarded a six-year contract for the industrial design of CIGEO. ANDRA expects to file a license application for the facility in 2015. Also by 2015,
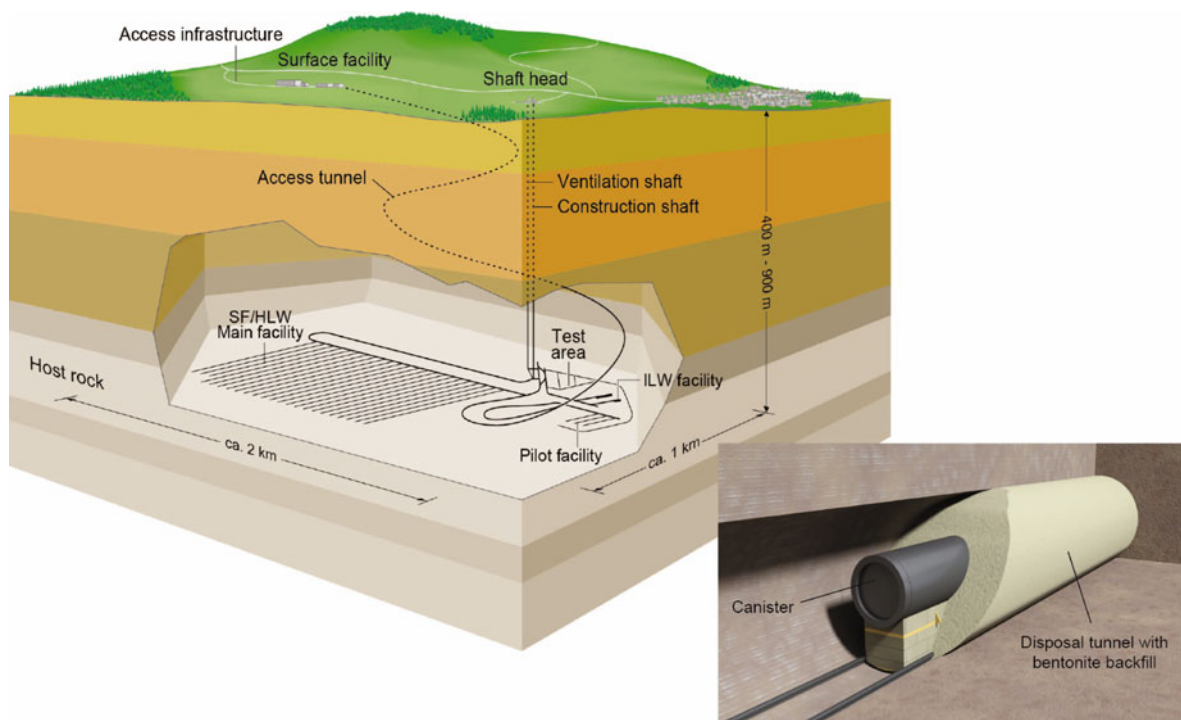
Figure 5. View of the Swiss concept for a SF/HLW/ILW repository in the Opalinus clay with inset of SF canister-emplacement tunnel and longitudinal section of emplacement tunnels for SF for the design option using low-pH shotcrete tunnel support. SF: spent fuel; HLW: high-level waste; ILW: intermediate-level waste (Copyright Nagra) [2002] (Nagra, 2002).

ANDRA is tasked with building or modifying storage facilities for H/ILW to meet future requirements.

In ANDRA's concept, a repository installation would consist of disposal cells (underground caverns) excavated in a claystone (Callovo-Oxfordian) formation; these cells contain the waste packages. The architecture envisioned by ANDRA includes disposal cells for various categories of waste within specific repository zones. An example of the layout of the repository zones for category B (ILW) waste can be found in Figures 6 and 7 (ANDRA, 2005).

The range of primary packages in terms of conditioning, geometry, and radiological and chemical content in the B waste category is highly diverse. The concern with simplifying operating methods has led to the design of standardized disposal packages which group together one to four primary packages in a parallelepipedal concrete container weighing ~6 to 25 tons and measuring from 1.2 to 3 m. The disposal cells are sub-horizontal tunnels limited in length to ~270 m. The concrete drift liner gives the engineered structure mechanical stability (ANDRA, 2005).

In the case of HLW, to prevent the inflow of water to the waste during the thermal phase, each primary package of vitrified waste is placed in a watertight overpack. This over-pack is made of non-alloy steel with an effective thickness of 55 mm, dimensioned very conservatively to withstand corrosion for 1000 y (ANDRA, 2005). The French concept does not rely on a bentonite buffer surrounding the waste packages.

However, bentonite seals are used to separate the disposal cells from the transfer drifts.

LONG-TERM PROCESSES – SCIENTIFIC UNDERSTANDING

To ensure that the safety functions described in the earlier section will be maintained for the repository timeframe, the long-term evolution of the repository must be evaluated. For this, an in-depth understanding of the processes acting on the barriers is needed.

The processes affecting the engineered clay barriers in a high-level waste repository are mainly due to the decay heat of the emplaced waste and to the perturbation of the host rock (formation of a disturbed zone, local desaturation of the host rock, and stress redistribution) caused by the excavation of the emplacement tunnels and caverns. The system is re-equilibrated by (thermally, hydraulically, mechanically, and chemically) coupled transient processes. These processes must be assessed carefully in order to prevent any permanent damage to the barriers. Processes affecting the bentonite buffer behavior in the near-field of a high-level waste repository include but are not limited to: heat transport; radiation-related processes; freezing; water uptake (unsaturated conditions); evolution of swelling pressure, including hydro-mechanical interaction with liner, host rock, and canister; pore-water flow; diffusive transport of solutes; gas dissolution and transport; bentonite interaction with metal corrosion products; bentonite interaction with cementitious
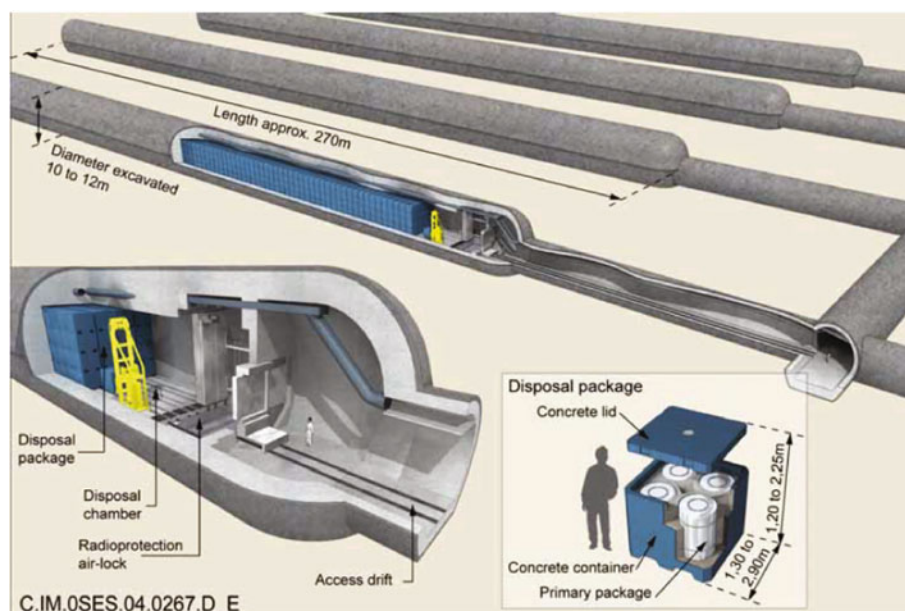
Figure 6. B-waste disposal cell in the French disposal concept.

materials; thermal alteration of bentonite; colloid generation/erosion; and microbial effects.

A brief overview of each of these processes will be given in the following sections.

Heat transport

Each high-level waste (or spent fuel) disposal canister produces ~1000−2000 W of heat by decay processes. This heat must be dissipated to limit the maximum temperature in the near-field and, thus, reduce the heat-related mineral alterations, and limit pore-water overpressure and dilatation of the rock matrix.

The maximum temperature in the bentonite buffer is reached at an early phase of the repository lifetime, after ~50 y, when the residual heat from the waste still is high and the bentonite is unsaturated or only partially saturated. At the interfaces between rock, bentonite, and canister, the unsaturated bentonite may leave small gaps through which heat can dissipate by radiation. The gaps are the required tolerances left from the installation
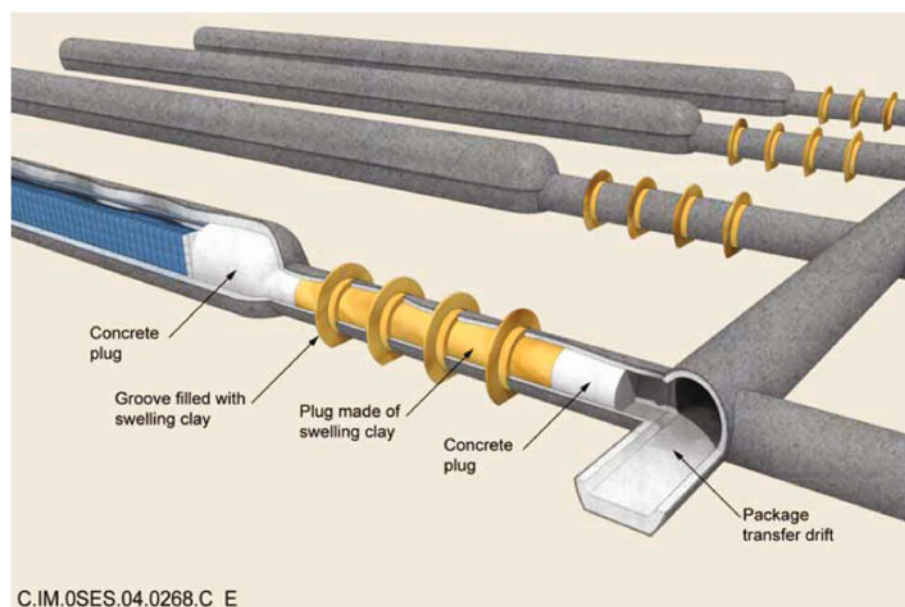

Figure 7. Sealed B-waste disposal cell in the French disposal concept.

of the canister in the bentonite. The presence of gaps has a substantial impact on the heat dissipation (Hökmark *et al.*, 2010). Heat dissipation by pure conduction across the bentonite near-field will also contribute to the initial heat dissipation. Convection may add to the heat transport, but due to the low permeability of the bentonite and because of the narrow gaps, this contribution is limited compared to conduction. When the buffer has become water-saturated, swelling is completed, and all gaps and joints are filled and heat is transported by conduction through the water-saturated bentonite. The efficiency of heat transport through the buffer region is important for the performance of the system, as it affects the maximum buffer temperature for which a design threshold has been established. Börgesson *et al.* (1994) measured the thermal conductivity of samples of compacted MX-80 bentonite by use of a heat-pulse technique (Figure 8). The results match well with corresponding results obtained for MX-80 by Plötze and Valter (2011) and with results obtained for similar materials, *e.g.* the bentonite tested in the FEBEX (Full Scale Engineered Barriers Experiment) experiment (Huertas *et al.*, 2000). The thermal conductivity of bentonite drops rather sharply at a degree of saturation of below ~50%, while it is fairly constant for a degree of saturation in the range ~50–100% (Börgesson *et al.*, 1994). Although, from a safety assessment viewpoint, there is no obvious need to define a lower boundary (critical value) for the thermal conductivity of bentonite, the bandwidth of data for thermal conductivity found in literature is taken into account when designing the near-field for high-level radioactive waste.

Radiation-related processes

The buffer will not be exposed to α- and β-radiation as long as the canister is intact. In the case of a failed canister, radionuclides that migrate out will be sorbed by the buffer and may expose it to a radiation dose. In calculations of radionuclide sorption, the total concentration of α-emitters in the buffer closest to the canister was found to be likely to give a total dose of 8×10^{15} alphas/g of bentonite for the first million years with the assumption of early canister failure. In most parts of the buffer, the α-dose will be much less as the α-emitters are strongly sorbed and will remain in the vicinity of the canister. 8×10^{15} alphas/g corresponds to an absorbed dose of 8 MGy. Gu *et al.* (2001) showed that the amorphization dose for montmorillonite is 30 GGy. As the total dose received is more than three orders of magnitude lower than the amorphization dose, the impact on the properties of the buffer can be expected to be very small. Exposure of the bentonite to γ-radiation from the waste could potentially damage the montmorillonite structure. This would lead to a decrease in the montmorillonite content in the bentonite and, thus, a reduction in swelling ability and an increased hydraulic permeability. However, experimental studies have shown that the accumulated radiation doses to which the bentonite will be exposed in a deep repository do not cause any measurable changes in the montmorillonite content (Pusch *et al.*, 1993). MX-80 bentonite saturated with weakly brackish water to a saturated density of 2050 kg/m^3 was irradiated for 1 y with a total radiation dose of 3×10^7 Gy, which is orders of magnitude more than will be the case in the repository. The specimen was confined in a cylinder with one closed end consisting of iron and irradiated with a ^{60}Co source, while the other end consisted of a porous steel filter through which a water pressure of 1.5 MPa was imposed (Pusch *et al.*, 1993). The irradiated end, which had an absorbed dose rate of 3972 Gy/h, was kept at 130ºC and the opposite end, which had a dose rate of 456 Gy/h, at 90ºC. Mineral assays (X-ray diffraction – XRD, infrared
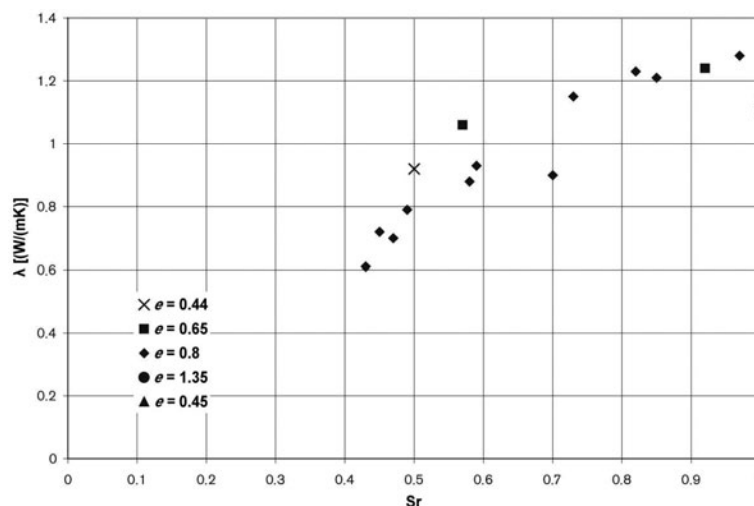

Figure 8. Heat conductivity (λ) of laboratory-scale bentonite samples as a function of saturation ratio (Sr) for a number of values of the void ratio (*e*) (from Börgesson *et al.*, 1994; reproduced with permission).

spectroscopy − IR, cation exchange capacity − CEC) of this specimen and of a parallel specimen not exposed to radiation but to the same hydrothermal environment showed no significant change in the montmorillonite quantity in either of the specimens.

Freezing

For repositories located in areas where the possibility of future permafrost conditions cannot be ruled out, the effect of bentonite freezing has to be considered. At temperatures below 0ºC, the water in the buffer can be anticipated to turn into ice. This is not an issue for the material itself as the process is reversible and it will regain its properties after thawing (Birgersson *et al.*, 2010). However, the formation of ice could lead to: (1) an increased pressure on the canister and rock; and (2) redistribution of material due to ice-lens formation.

Birgersson *et al.* (2010) investigated the behavior of compacted bentonite below 0ºC. How swelling pressure (*i.e.* sealing properties) changes with temperature and under what conditions bentonite freezes was investigated. The freezing point of a soil sample is defined as the temperature at which ice starts to form in the material. When ice formation occurs in confined bentonite, a substantial pressure increase is expected due to volume expansion of water. The swelling properties of bentonite make it unique in the sense that its properties depend on external conditions. The process of swelling, specifically, only occurs when bentonite is in contact with an external aqueous reservoir, and the concept of swelling pressure can consequently only be defined under such conditions. This means that a bentonite component within a repository will be affected by freezing as soon as the groundwater in the surrounding rock freezes, even though the component itself may

remain unfrozen. The process of freezing bentonite, therefore, also includes the temperature range between the freezing point of the aqueous reservoir and the actual freezing point of the bentonite.

A critical temperature occurs somewhere below 0ºC, denoted here as T_c and measured in ºC, at which swelling pressure is completely lost. T_c depends only on swelling pressure measured at 0ºC, and not explicitly on clay-specific quantities such as montmorillonite content, montmorillonite layer charge, or density.

A theoretical description of the pressure response due to temperature in bentonite above T_c was provided by Birgersson *et al.* (2008, 2010). These results demonstrate that any bentonite component of the KBS-3 repository will strive to lower its pressure as the temperature drops below 0ºC. The size of the equilibrium-pressure drop depends essentially, for all relevant bentonite densities, on the difference in molar entropy between bulk water and ice only, which at 0ºC corresponds to 1.2 MPa/ºC. Because the pressure drop is determined by properties not related to the clay, the same behavior is expected independent of the specific bentonite being used. Pressures have been evaluated for samples of the Ibeco RWC bentonite for temperatures both below and above 0ºC (Figure 9). A typical bentonite buffer will have a freezing point of ~−6ºC or lower, which is considerably lower than what can be expected at repository depth at any considered repository location, even in countries like Sweden and Finland where permafrost conditions are expected for a substantial part of the repository evolution.

Water uptake (unsaturated conditions)

The bentonite buffer is installed as partially saturated blocks and pellets. The initial degree of saturation is dependent on the particular concept. The safety func-
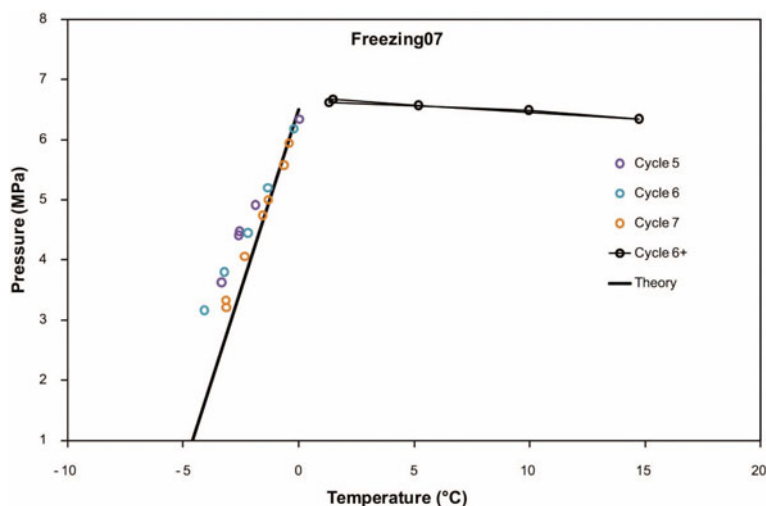

Figure 9. Equilibrium pressure *vs*. temperature for Dep-Can (Ibeco RWC) bentonite at a swelling pressure of ~7 MPa at temperatures >0ºC. The freezing point (T_c) of the sample is below −8ºC. The line labeled 'Theory' shows the expected pressure response if the water-saturated bentonite is treated as thermodynamically equivalent to a salt solution (from Birgersson *et al.*, 2010; reproduced with permission).

tions for the buffer and backfill assume a fully water-saturated state. This means that the buffer and backfill need to be saturated to perform properly. However, no performance is required from the buffer as long as the deposition hole is unsaturated, as no mass transfer of dissolved species (corrodants or radionuclides) between the canister and the groundwater in the rock can take place in the unsaturated stage. The water saturation process itself, therefore, has no direct impact on the safety functions of the buffer and backfill. The repository layout is determined for an unsaturated buffer, which means that the maximum temperatures allowed will not be exceeded even in the absence of water uptake. An understanding of the water-saturation process is still needed as it defines the state of the barriers in the early evolution of the repository. Finally, the ventilation of deposition tunnels in clay rock during significant time spans (prior to filling of the deposition holes and the tunnels) may cause the surrounding rock to dehydrate. To a certain extent, the host rock will desaturate during the excavation phase and slowly resaturate along with the emplaced dry bentonite pellets. High suction pressures and osmotic effects drive the water uptake in the near-field.

The uptake of water in compacted bentonite has been studied extensively, both in laboratory experiments and in large-scale field tests. One example of a full-scale test is the FEBEX test in the Grimsel underground laboratory in Switzerland (Huertas *et al.*, 2000).

Evolution of swelling pressures, including hydro-mechanical interaction with liner, host rock, and canister

Water is adsorbed by both unsaturated and saturated bentonite (that is able to physically expand) and causes swelling (Norrish, 1954). If the bentonite is unable to expand freely, a swelling pressure develops, which reaches its peak at full water saturation.

The swelling pressure is a key safety function of the repository. The swelling ability sets limits on microbial activity by reducing macro porosity, keeps the canister in position, prevents any further propagation of the excavation disturbed zone (EDZ) in the rock, reduces the hydraulic conductivity, takes up shear forces from the rock, and ensures that the buffer is self-sealing.

Understanding of the swelling process is, therefore, critical in the evaluation of the long-term performance of a bentonite buffer. The swelling pressure is strongly coupled to the density of the material and a relatively small change in density can lead to significant change in swelling pressure. The swelling pressure for a given bentonite (Figure 10) is not very sensitive to either the counter-ionic or ionic strength of the water at the design dry density for a repository ($1500-1600$ kg/m^3) (Karnland *et al.*, 2006). This picture changes at low density, where both counter-ionic and ionic strength have a large impact on the swelling pressure.

One important aspect to study is the homogenization, or actually the incomplete homogenization, of the installed material. In most design concepts, the near-field will be filled with a mixture of pellets and blocks used to support the canister. The use of different forms of materials with different densities and degrees of compaction will lead to density differences and swelling-pressure differences in the buffer, mainly due to friction within the material, but also due to hysteresis effects. The remaining inhomogeneities in the buffer material have been studied using different approaches (Komine and Ogata, 1994; Komine and Ogata, 2004; Sridharan, 1997; Villar, 2007; Wang *et al.*, 2012).

One example is the full-scale Canister Retrieval Test (CRT) at the Äspö Hard Rock Laboratory in Sweden. The test, which simulated a deposition hole with canister and buffer material according to the design of a KBS-3 deposition hole (Figure 1), was run for >5 y and excavated with extensive sampling and determination of the density and water ratio of the buffer material.

The outside diameter of the canister was 1050 mm, the height was 4830 mm, and the total weight was 21.4 metric tons. The bentonite used as the buffer material was Wyoming bentonite (MX-80). The buffer was installed as highly compacted bentonite blocks and rings with an initial bulk dry density of 1710 and 1790 kg/m^3, respectively. The initial water content of the bentonite was 17% by mass. The space between the bentonite blocks and the borehole wall was filled with bentonite pellets and water. The test has been subjected to extensive modeling by many different teams. Examples of results can be found in Zandarin *et al.* (2012), where the degree of saturation and the dry density at the end of the test have been calculated and compared to the values measured after excavation (Figure 11), and in Kristensson and Åkesson (2011), where an analytical model and a finite element solver were applied and both were capable of predicting the final average void ratio of the pellets and the blocks in the experiment.

In a sedimentary host rock the formation, propagation, and sealing of the EDZ must be carefully assessed as it may cause undesirable networks of higher permeabilities along tunnel axes. The EDZ is understood to form by and during the excavation of underground tunnels and drifts and is mainly caused by stress redistribution. The formation and propagation of the EDZ can be limited by supporting the tunnel walls by reinforcement using cement lining, rock bolts, and other engineering solutions and using adapted excavation methods such as roadheaders. In large-scale experiments (*e.g.* SELFRAC-I experiment) swelling pressures of bentonite simulated by a plate load of ~4.8 MPa were shown to reduce the transmissivity of the EDZ by several orders of magnitude to values of only one to two orders of magnitude greater than in pristine Opalinus Clay (Bock *et al.*, 2010).
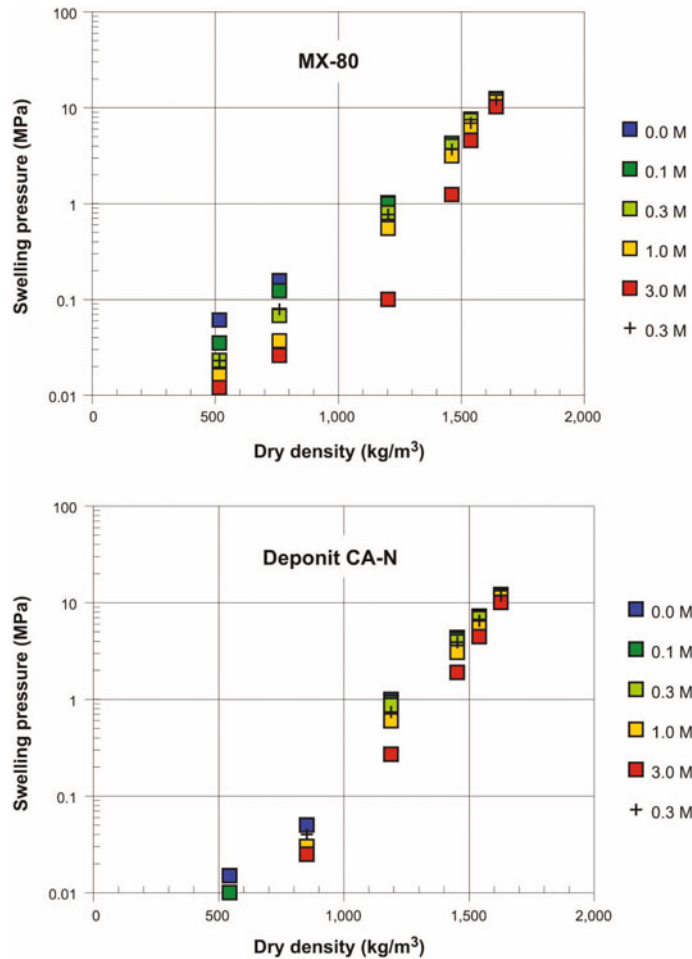
Figure 10. Swelling pressures of MX-80 exposed to NaCl solutions (upper) and Ibeco RWC (Deponit CA-N) exposed to CaCl₂ solutions (lower). Concentration expressed as M (from Karnland *et al.*, 2006; reproduced with permission).

Pore-water flow

Together with the self-sealing ability, the low hydraulic permeability under saturated conditions is the most important property of bentonite for nuclear-waste applications. For a buffer in a high-level waste-disposal concept, the low permeability (usually expressed as hydraulic conductivity) ensures that diffusion is the dominant transport mechanism and advection can be ignored for a high-performing buffer. Maintaining the low conductivity in the repository is, therefore, important. Recently, some debate has focused on how the hydraulic conductivity of bentonite is affected by heating and how hydraulic conductivity should be measured. Pusch *et al.* (2010) reported that the hydraulic conductivity of the MX-80 bentonite used in the CRT test (mentioned in the previous section) increased by three orders of magnitude after 5 y of heating up to 95ºC. No record of how the hydraulic testing was performed was given. The same sample showed no change in swelling pressure. The authors still interpreted this as a change in clay mineralogy. More

careful and systematic testing of samples for the same test performed by Dueck *et al.* (2011) showed no significant changes in either hydraulic conductivity or clay mineralogy compared to reference samples.

Harrington *et al.* (2014) also studied the hydraulic conductivity of samples from the same test. They decided to inject distilled water at a flow rate of only 1.6 µL/h in order to minimize any perturbation of the system and avoid the potential movement of dissociated mineral components (if any exist). If the bentonite sample actually exhibited a conductivity of 2.0×10^{-11} m/s as reported by Pusch *et al.* (2010), then this would generate a head gradient of only 8 m/m. However, the pressure gradient required to achieve this flow was in excess of 3000 m/m and not the 8 m/m as predicted, yielding an average hydraulic conductivity of 4.7×10^{-14} m/s. This value is in close agreement with that expected for unaltered material and is also consistent with the measurements of Dueck *et al.* (2011). This shows clearly that suspicious measurements of hydraulic conductivity should be treated with great
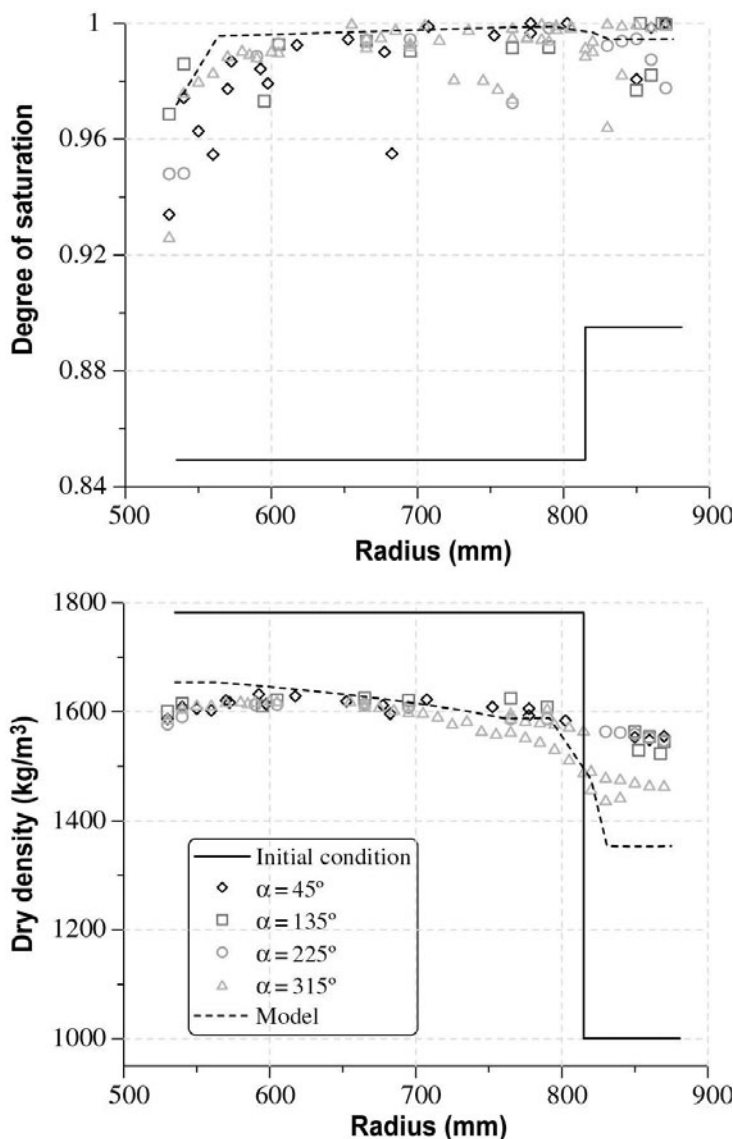
Figure 11. Degree of saturation and dry density for a buffer ring at canister mid-height after test dismantling. Experimental and model results (from Zandarin *et al.*, 2012; reproduced with the permission of Elsevier).

caution and that bentonite does not change its properties after a 5 y heating period.

Diffusive transport of solutes

As stated in the previous section, the low hydraulic conductivity of the bentonite buffer together with the low hydraulic gradient of the near-field and the host rock will ensure that nuclide transport is limited to slow diffusive transport. Different elements will, however, be exposed to different conditions within the bentonite. The permanent negative charge of the clay will impose restrictions on the movement of anions. This has been interpreted with different conceptual descriptions. One approach is to distinguish between different types of pore water: inter-layer water and inter-particle water

subdivided into external free water and water associated with the diffuse double layer (Wersin, 2004; Appelo, 2011), where diffusion of anions is restricted to the free water. The other approach assumes that virtually all water is localized in the interlayer due to the large surface area of the clay and the high density of the buffer (Birgersson and Karnland, 2009). This approach uses the Donnan equilibrium between the buffer and the external water in the host rock to account for the reduced transport capacity of anions. Despite the theoretical differences, both approaches have been able to satisfactorily model the diffusive transport for both cations and anions and the selection of the approach may, therefore, not be an important issue for the purpose of modeling long-term performance.

Another key feature involved in the diffusive transport of species in the bentonite buffer is the strong sorption of many elements. Sorption can either take place at the permanently charged layer surface of the clay mineral or at the variably charged edge surface. On the layer surface, sorption takes place when charge-compensating ions are exchanged. Sorption on the edges takes place due to the formation of surface complexes. Ion-exchange is the typical sorption mechanism for alkali and alkaline-earth elements, as well as transition metals at low pH values where positive species are predominant. Surface complexation is generally the more relevant process for all reactive elements (transition metals, actinides, lanthanides, and reactive anions such as carbonate). Both ion-exchange and surface complexation take place simultaneously, but at different surfaces. Sorption of radioelements onto bentonite has been studied extensively by a large number of laboratories. A few examples of this are: Grambow *et al.* (2006), who studied sorption of cesium, nickel, lead, and some trivalent nuclides on MX-80 bentonite; Bradbury and Baeyens (2005), who supplied new data for U(VI) sorption on bentonite; Missana *et al.* (2009), who provided new data for Se(IV) sorption; and Huber *et al.* (2011), who studied the reversibility of sorption of radionuclides on bentonite colloids.

Gas dissolution and transport

Gas will be produced by corrosion of the waste canister and, to a limited extent, by the waste matrix itself. The prevailing gas-transport mechanisms are advection and diffusion of dissolved gas, two-phase flow, and pathway dilation (Marschall *et al.*, 2005). This is especially true for partially saturated argillaceous media. Gas generation through corrosion and degradation processes in the backfilled tunnels and galleries might lead to gas overpressures in the engineered barriers and host rock and to some displacement of pore-water around the repository. In an unsaturated or partially saturated bentonite, a linear dependence develops between the gas-flow rate and pressure gradient, which indicates that two-phase flow is the dominant transport mechanism (Villar *et al.*, 2012). This may also be the case for saturated sand-bentonite mixtures if the sand content is sufficiently high. At a degree of saturation of ~80−90% or higher, the behavior changes entirely (Villar *et al.*, 2012). No flow of gas will take place in the bentonite unless the applied pressure is equal to or greater than the total stress (swelling pressure + back pressure). The only transport mechanism is the omnipresent diffusion of dissolved gas. If the applied gas pressure reaches a higher value than the total pressure in the bentonite, a mechanical interaction between the gas and the clay will occur. This interaction will lead to either consolidation of the bentonite or formation of dilatant pathways. Consolidation means that a gas volume will be formed within the clay and the

clay will be compressed. This increases the clay density closest to the gas volume and the local swelling pressure is increased to balance the gas pressure. The extent of consolidation is, however, limited. At some critical pressure, pathways will be formed and the gas will become mobile. The pathways are characterized by a strong coupling among the factors of total stress, swelling pressure, and pore pressure; localized changes in total stress, swelling pressure, and pore pressures; and unstable flow, exhibiting spatio-temporal evolution, localized outflows during gas breakthrough, and no measurable desaturation in any test samples (Graham *et al.*, 2012). The transition between the end of consolidation and the start of pathway formation is still unclear. In some tests, pathways form when the gas pressure reaches the total stress in the sample. An example of this is the large-scale gas-injection test (Lasgit) in the Äspö rock laboratory (Cuss *et al.*, 2011).

Bentonite interaction with metal corrosion products

Most of the concepts for radioactive-waste disposal foresee built-ins made of steel (carbon or non-alloy steel) or even steel canisters (*e.g.* Switzerland and France). Under both aerobic (*e.g.* construction phase) and anaerobic conditions (*e.g.* operational phase), steel will corrode and form corrosion products. The corrosion rate of iron in a compacted clay will be in the range of ~1 μm/y or less (Johnson and King, 2008). Copper is generally stable in a repository environment, except for the reaction with aqueous sulfide to form solid copper sulfides. No interaction between copper ions and bentonite is expected as the solubilities of copper sulfides and copper metal are too small to reach a significant concentration of dissolved copper. Uncertainties remain as to how far corrosion products such as H_2 gas or Fe^{2+} cations will interact with the clay (*e.g.* reduction of octahedral Fe^{3+} in bentonite) and alter its safety-relevant properties (*e.g.* swelling capacity, nuclide retention capacity, *etc.*) or form new Fe-rich minerals (*e.g.* berthierine, odinite, greenalite, *etc.*) (Bradbury, 2012).

Bentonite interaction with cementitious materials

The slow resaturation process of the near-field will allow for highly coupled and non-linear reactions (Figure 12) between cement construction materials, particularly the liner, and clay minerals (bentonite and clay host rock). This could lead to various alterations of the clay, which might have a negative effect locally on the safety-relevant properties of the barriers, such as hydraulic conductivity, gas permeability, swelling capacity retention of nuclides, *etc.*

Precipitation and dissolution of clay minerals will alter the transport properties of the clay barriers and, thus, affect the spatial extent and the timescale of these alteration processes. Depending on the type of cement, these precipitation and dissolution reactions together
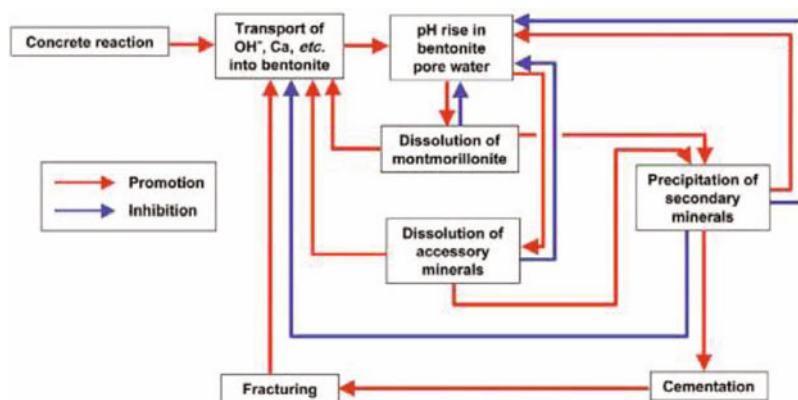
Figure 12. Cement-bentonite interaction as a coupled non-linear system (Copyright Nagra) [2013].

with tunnel convergence or other non-isotropic loads might lead to weakening of the structural strength of the cement and result in (micro-) fractures accelerating the degradation of the cement. In addition, the heat pulse from the radioactive decay might also influence the kinetics of these reactions.

Reactions between concrete and clay are generally slow, which makes them difficult to study experimentally. The non-linearity makes extrapolation to repository timescales difficult. One example of an experimental study was presented by Yamaguchi *et al.* (2007), where compacted discs of bentonite/sand mixtures were exposed to NaCl-NaOH solutions of variable pH at elevated temperatures. The study showed a different pH dependence of the montmorillonite dissolution rate compared to results from studies with loose materials (*e.g.* Huertas *et al.*, 2001; Sato *et al.*, 2004). This indicates that reactions may be slow in compacted clay. However, due to the large uncertainties, the preferred option is usually to design liners and plugs to be constructed from a concrete that would give a 'low' pH pore water (pH <11).

Thermal alteration of bentonite

The advantageous physical properties of the buffer, principally swelling pressure and low hydraulic conductivity, are determined by the capacity for water uptake between the montmorillonite layers (swelling) in the bentonite. Transformation from montmorillonite to illite, which is the most common alteration observed in natural sediments (*e.g.* Burst, 1959; Hower and Mowatt, 1976), is well documented in different geological environments (*e.g.* Jennings and Thompson, 1986; Horton, 1985) and has been reproduced under laboratory conditions (*e.g.* Eberl, 1978; Güven and Huang, 1991). The conversion always involves a layer-charge increase, mainly due to a decrease in silica content, and an uptake of charge-compensating K^+ ions. A large amount of smectite is commonly found in old formations exposed to repository-relevant temperatures. For example, the time-temperature space of illitization in seven deep wells in four sedimentary basins in the US, Japan, and France was studied by Velde and Vasseur (1992). In all wells, a typical reduction of smectite content with depth was observed, which represents an increase in both age and temperature (Figure 13).

The prerequisites for a transformation are obviously present in the sediments, and time and temperature are the governing parameters. A decrease to ~60% smectite was observed in the Californian Norwal formation after 4.5 My at a depth of 5 km, representing a final temperature increase of >100ºC. The same transformation took ~60 My at a depth of 2 km and a temperature increase of ~70ºC in the Texan Peeler sediments. The reaction rate at these repository-relevant temperatures is consequently very slow in relation to the timescale of a repository. Illitization requires K, which may be transport limited in the repository environment. The bedrock, however, consists partly of K-feldspar, which means that K is available in the vicinity.

Kinetic models have been developed for the smectite-to-illite transformation. Relationships and constants have been established by means of laboratory batch experiments and comparisons with natural systems (*e.g.* Eberl and Hower, 1976; Pytte, 1982; Velde and Vasseur, 1992; Huang *et al.*, 1993).

Most models take the form of an Arrhenius expression. While kinetic models can be used to explain observations, their use in long-term predictions over timescales up to 1 My may be somewhat speculative. For this particular process, the comparison with natural observations gives a more powerful argument. However, the kinetic models are still valuable as they can be used to define the maximum temperature allowed in a repository.

Colloid generation/erosion

Swelling of the bentonite buffer as a result of uptake of water is counteracted by the walls of the deposition hole and a swelling pressure is developed in the bentonite. Fractures intersecting the deposition hole
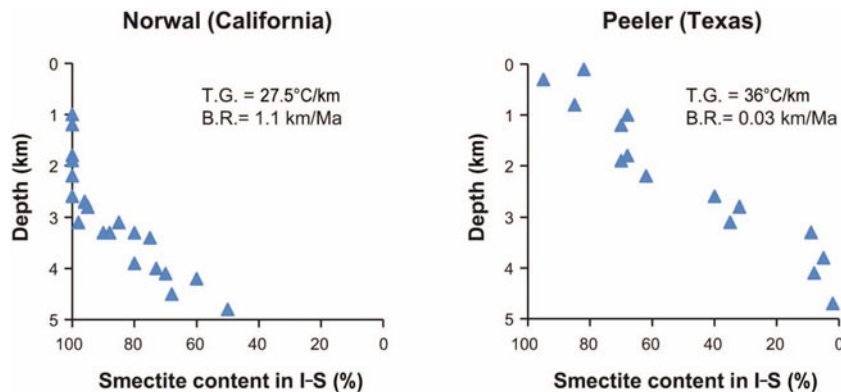
Figure 13. Smectite content *vs.* depth in smectite-illite mixed-layer material in two sediments representing relatively fast burial rate (left) and slow burial rate (right). T.G. indicates the present temperature gradient and B.R. indicates the burial rate (after Velde and Vasseur, 1992; reproduced with permission).

mean that rigid swelling restrictions are not present everywhere and that localized swelling continues into the fractures until an equilibrium or steady state is reached. This free swelling may lead to separation of individual montmorillonite layers (dispersion) and part of the buffer could be transported away by groundwater.

The maximum free swelling of the bentonite is strongly dependent on the valence and concentration of the ions in the interlayer space (Norrish, 1954). At low concentrations of dissolved ions in the groundwater, the interlayer distance between the individual montmorillonite layers may increase sufficiently to give the clay/water system a sol character, *i.e.* single or small groups of montmorillonite layers act as individual colloidal particles.

Non-saline waters may solubilize the smectite particles in the bentonite and carry them away as colloidal particles. This is known to happen if the water content of ions is below the critical coagulation concentration (CCC). Above the CCC, the smectite forms a coherent gel, which does not release particles.

In a situation when colloids are released, the mass of the bentonite barrier will decrease. This may eventually lead to a situation where expected and desired diffusion control is lost. A loss of the diffusion barrier in the buffer in a fractured rock will lead to a significant increase in canister corrosion and/or radionuclide release from the repository.

A CCC may be determined for monovalent systems and used as a pessimistic concentration limit for spontaneous colloid particle release (*e.g.* Evans and Wennerström, 1999). The governing variables are the concentration and the layer charge of the montmorillonite. Laboratory results have shown that montmorillonite extracted from MX-80 bentonite and ion-exchanged to a Na^+-state has a CCC of ~25 mM in NaCl solution, *i.e.* an attractive gel is formed (Birgersson *et al.*, 2010). Two other Na-montmorillonites analyzed with a higher layer charge than in MX-80 were found to have a lower CCC, in contrast to predictions from the DLVO theory for parallel layers (Evans and Wennerström, 1999). One

possible explanation for this is that the clay with the lowest CCC had a considerable amount of tetrahedral charge, which should give a larger edge−face attraction (Hetzel and Doner, 1993). For systems with only divalent counter-ions, the CCC concept is not really valid and the CCC may be considered to be zero, *i.e.* no excess ions are needed in order to prevent colloidal sol formation of colloidal particles.

A DLVO-based force-balance model for spherical colloids (Petsev *et al.*, 1993) has been adapted for parallel clay layers (Liu *et al.*, 2009; Neretnieks *et al.*, 2009) and used to calculate the swelling of Na-montmorillonite into fractures filled with water of low ionic strength. The force-balance model uses DLVO to describe swelling pressure and a Kozeny-Karman-like expression fitted to experiments to describe hydraulic conductivity. Advective loss of montmorillonite is modeled by combining the force-balance model for swelling with a viscosity model for the repulsive montmorillonite gel and the Darcy equation for two-dimensional flow in a fracture intersecting the deposition hole. The outcome of the modeling for different water velocities in a 1 mm aperture fracture is shown in Table 1. The montmorillonite release rate is found to be proportional to the water velocity to the power of 0.41 and directly proportional to the aperture.

Colloids bearing radionuclides may be formed during SF/HLW dissolution or subsequently by precipitation of insoluble radioelements or by co-precipitation and sorption processes. The small pore size in dense, saturated bentonite, however, prevents transport of colloids (Kurosawa *et al.*, 1997). Bentonite is, therefore, an effective transport barrier for radionuclides.

Microbial effects

Microbial activity in the Opalinus clay and in compacted bentonite with a dry density >1.45 g/cm^3 is considered to be low due to the small size of the pores, their poor interconnectivity and the restricted availability of water (Stroes-Gascoyne, 2002, 2011).

Table 1. Loss of smectite by advective flow (from Neretnieks *et al.*, 2009; reproduced with permission).

Water velocity (m/y)	Smectite release for a 1 mm fracture aperture (g/y)	Penetration into the fracture at the center (m)
0.10	11	34.6
0.32	16	18.5
0.95	26	11.5
3.15	42	7.0
31.5	117	2.0
315	292	0.5

Nevertheless, a catalytic influence of microbes on redox processes is conceivable.

One microbial issue that may require more attention in the future within the radioactive-waste management community is the role of microbes in the smectite-to-illite reaction. Microbes can promote the smectite-to-illite reaction at room temperature within 14 days (Kim *et al.*, 2004). This contradicts the evidence from natural systems presented above. The questions then are: under what conditions can microbes affect the reaction and do these conditions prevail in an underground repository? The activity of microbes will be limited in a dense clay, which means that the process should not occur within bentonite blocks or in interfaces between bentonite and a clay host rock. However, reactions in the interfaces between bentonite and other materials (*e.g.* canister) and host rocks, as well as in low-density pellets, are still a concern.

ENGINEERING ASPECTS

Sweden

The reference design of the buffer in the Swedish KBS-3 system (Figure 1) is described by a set of design parameters for which nominal values and acceptable variations are given. The design parameters will be inspected in the production to confirm that the buffer produced in the initial state conforms to the reference design and to provide an estimate of the actual properties of the buffer in the initial state. The reference design of the buffer consists of specifications of the material composition, the material ready for compaction, the fabricated blocks and pellets, and the buffer installed (Figure 14). The buffer consists of one solid bottom block, six ring-shaped blocks around the canister, and three solid blocks on top of the canister. The buffer ends and the backfill commences at the top of the third block on top of the canister. The center line of the buffer blocks coincides with the center line of the deposition hole. The gap between the blocks and the rock surface of the deposition hole is filled with pellets.

The production of the buffer consists of three main parts: excavation and delivery, manufacturing of blocks and pellets, and handling and installation.

Bentonite deposits exist in many places around the world and excavation and delivery can be provided by a range of companies. The desired material properties will
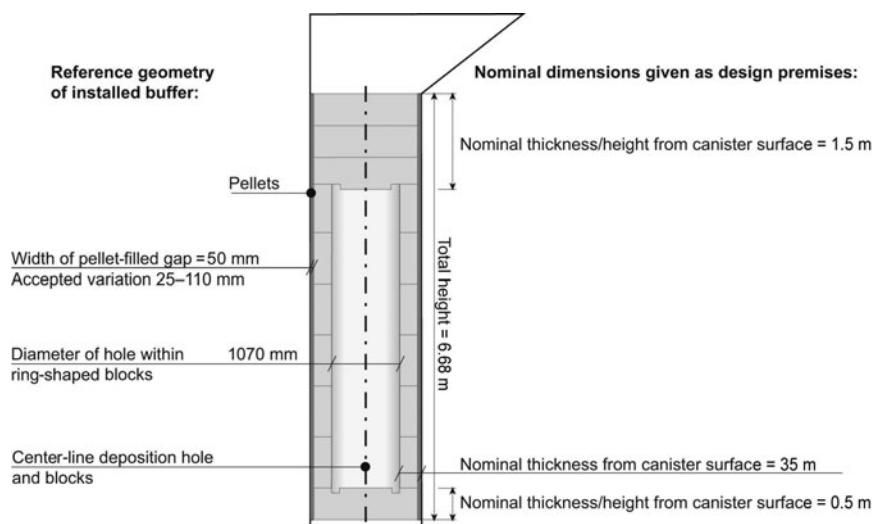

Figure 14. Reference geometry of the installed buffer in the Swedish concept.

be specified upon ordering. Each shipment of bentonite will be accompanied by a protocol from the supplier that describes the actual composition of the material delivered. The material delivered is inspected as the basis for the acceptance of the delivery. Inspections for the production comprise measurement of water content and granule size distribution of the delivered material and measurement of its total weight. Design parameters of importance for the functions of the buffer to be inspected include the mineralogical composition of the buffer material. The mineralogical composition of the bulk material is analyzed as random powders using a standard X-ray diffractometer. This will be supported by CEC determinations, swelling pressure, and hydraulic conductivity tests, *etc.* Before compaction, the water content of the material is adjusted to suitable values for the process. The reference method for pressing of blocks is uniaxial compaction. After compaction, the weight and dimensions (height and (inner and) outer diameter) of each block are inspected. The dry density of the block is calculated from the recorded water content, weight, and dimensions. The blocks are machined to specified dimensions in order to achieve a well aligned stack of blocks in the deposition hole and the required installed bulk density. The reference method for manufacturing pellets is to compact the conditioned material to small pellets.

To protect the buffer from taking up water from the moisture in the air and from drying if the deposition hole is dry, a protective sheet is placed in the deposition hole before installation of the blocks. The bottom block is installed and centered with respect to the average center line of the deposition hole. The first ring-shaped block is installed in the same way as the bottom block. The installed ring-shaped block is used to guide the installation of the next ring-shaped block so that they are positioned in the same horizontal position on top of each other with a straight and centered hole for deposition of the canister in the middle. When the canister has been deposited, the top three buffer blocks and the two blocks considered as a part of the backfill are installed. The pellets are used to fill the gap by placing a conical hood on top of the last bentonite block to be installed and pouring the pellets into the deposition hole. As soon as the gap has been filled with pellets, the backfilling of the deposition tunnel continues so that the backfill can prevent swelling and expansion of the buffer caused by a fast water uptake of the pellet filling.

The techniques for loading, transportation, and storage of the buffer blocks and pellets are well known from similar industrial applications. SKB has relevant experience from the Äspö HRL. Blocks have been stored on pallets with an air-tight hood for several months without showing any changes in weight or water content.

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Apart from properties intrinsic to clay (resistance to mineral transformation, nuclide retention, *etc.*), the safety-relevant properties of the bentonite buffer and backfill are affected by the water content and the emplacement density. Thus, the fabrication and the emplacement of the bentonite elements such as blocks, pellets, and chips will define how the buffer and backfill will perform over the timespan considered for the safety analyses. Considerable effort is, therefore, invested in the development of emplacement techniques and production procedures for high-quality bentonite blocks, pellets, chips, and granulates to fulfil the safety criteria.

Large-scale demonstration experiments such as the 'Prototype repository experiment' at the Äspö underground laboratory or the 'Full-scale emplacement' experiment at the Mont Terri underground rock laboratory have been instrumental in verifying the performance of buffer and backfill under field conditions and in testing emplacement techniques.

In NAGRA's concept, the waste canisters are placed on a pedestal made of bentonite blocks. These blocks are designed to support the weight of a single canister which might reach up to 25 t, which translates into a pressure of ~0.1 MPa.

The emplacement tunnels are to be backfilled with a mixture of bentonite granulate, chips, and pellets. The mixture of the three different grain sizes is chosen to fit a Fuller curve in order to achieve an homogeneous distribution of the density in the near-field. The emplacement density of this mixture ensures that the hydraulic conductivity remains low enough to ensure diffusive transport, the gas permeability is high enough to allow for gas transport without plastic deformation of the buffer or sedimentary host rock, the porosity is sufficiently low to inhibit bacterial growth, and the density is sufficiently high to seal the EDZ by swelling and to limit colloidal transport.

CONCLUSION

Bentonite clay and bentonite clay mixtures are used as buffers, seals, or backfills in almost every program for radioactive-waste disposal worldwide. The development of the concepts started in the late 1970s and extensive research and technology development has been going on since then. The key features of bentonite in this application are low hydraulic permeability, self-sealing ability due to the swelling pressure, and long-term mineralogical stability. The great functionality of bentonite has been demonstrated in a large spectrum of laboratory and *in situ* experiments. In a crystalline, fractured host rock below the groundwater table, practically no alternatives to a bentonite buffer are possible. In a clay host rock, the geological medium itself acts as a self-sealing hydraulic barrier and other options may be possible (*e.g.* back-filling with host rock or using a concrete barrier), but a clay buffer may also have advantages. Bentonite is still the main option for seals in shafts and galleries, both in crystalline and clay host rocks.

In both Finland and Sweden, license applications have been filed for the actual construction of repositories, based partly on an engineered barrier constructed out of bentonite. Future research will still be needed, however, to further confirm the performance of bentonite barriers. A few areas currently of interest are: effects on bentonite of long periods of unsaturated conditions together with elevated temperatures; redistribution of mass during swelling, which will impact how pellets and blocks of different initial density will interact and how potential losses of material can be resealed; further understanding of the colloid formation/erosion processes − in Sweden and Finland, periods with dilute water after melting of future glaciers cannot be excluded and a large loss of bentonite may have a direct impact on the overall safety of a repository; stability of bentonite under non-isothermal conditions and different degrees of saturation; and redox stability of bentonite under highly reducing conditions (saturation of the near-field with hydrogen gas) and its impact on the safety-relevant properties of bentonite.

National legislation also requires regularly renewed long-term safety assessments even if a repository license is granted. Consequently, research will continue in order to minimize uncertainties in the evolution of material properties (of bentonite) in the context of a near-field of a high-level waste repository.

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