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D4.1 Integration of the Short-term Evolution of the Engineered Barrier System (EBS) with the Long-term Safety Perspective

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1 Introduction

1.1 The PEBS Project

The main aim of the project PEBS (Long-term Performance of the Engineered Barrier System) is to evaluate the sealing and barrier performance of the EBS with time, through development of a comprehensive approach involving experiments, model development and consideration of the potential impacts on long-term safety functions. The experiments and models cover the full range of conditions from initial emplacement of wastes (high heat generation and EBS re-saturation) through to later stage establishment of near steady-state conditions, i.e. full re-saturation and thermal equilibrium with the host rock. The intention is to integrate the knowledge obtained in the project in a manner that will lead to a more convincing connection between the initial transient state of the bentonite barrier and its long-term state that provides the required isolation of the wastes. The work within the project builds on existing knowledge and experience generated during recent years and supported by ongoing national and EC research programmes. The project intends to provide a more complete description of the THM and THM-C (thermo-hydro-mechanical-chemical) evolution of the EBS system, a more quantitative basis for relating the evolutionary behaviour to the safety functions of the system and a further clarification of the significance of residual uncertainties for long-term performance assessment.

It is noted that there are other phenomena relevant to the engineered barrier system that have not been dealt with in the PEBS project. Gas production and transport processes in bentonite barriers have been studied in some detail in the FORGE project (Shaw 2014). In addition, ongoing activities within the IGD-TP (Implementation of Geological Disposal Technology Platform) are examining microbial issues relevant to geological disposal.

1.2 Objectives of D4.1

The present report focuses in particular on the importance of uncertainties arising from potential disagreement between the process models for THMC evolution and the relevant laboratory and in situ experiments performed both within and outside PEBS. The implications for extrapolation of results are treated, with particular emphasis on extrapolation of long-term performance. The report builds on the approach presented in PEBS Deliverable 1.1 (SKB et al. 2012), which described the treatment of the early evolution of the EBS in safety assessments from a number of European national programs based on their studies published in the period 2002 - 2010. Despite the differences in repository concepts, which include crystalline, and clay host rocks and copper and carbon steel canisters, the safety functions defined for the engineered clay barriers are similar. The key processes occurring in the EBS in the early evolution of the repository that may affect the long-term performance are similar in all concepts on a fundamental level. However, the significance of the processes for long-term safety as well as the treatment of the processes in the safety assessment can differ among the different concepts.

In Deliverable 1.1, the safety assessment methodology was discussed with particular emphasis on **safety functions**, which are a tool that is used for the evaluation of the long-term performance of individual repository components. The safety function approach as applied to a bentonite barrier is reiterated here in Chapter 2 to provide a framework for the subsequent discussion, as the use of safety function indicators arises frequently throughout the report in the discussion of the significance of uncertainties for long-term performance.

The main body of the present report describes the performance of the bentonite barrier in the context of four "cases" that were defined at the outset of the project in D1.1. These cases represent a method of examining the various THMC processes that occur during the evolution of the EBS, along with their impacts on long-term performance, in a manner that provides for each case a specifically different focus. The four cases are outlined in Chapter 3, including the reasons for their selection and the main work in the project that contributes to evaluating them. The four defined cases provide a good basis for discussing all aspects of evolution of the EBS, which forms the content of Chapter 4 and focuses on the best understanding of the processes and the associated uncertainties.

Chapter 5 uses the safety function approach to elaborate the importance of the derived uncertainties in process understanding in terms of assessment of the long-term performance of the bentonite barrier, focusing in particular on the degree to which hydraulic and swelling properties are likely to be affected over the long term as a consequence of the early thermo-hydraulic evolution phase. However, because the significance of the uncertainties in terms of disposal system performance depends on other considerations as well, in particular the specific repository design concepts and the barrier functions of the host rock and canister, the consequences of the uncertainties are discussed Chapter 5 in the context of the national safety cases.

2 General Discussion of Safety Functions, Important Processes and Key Identified Uncertainties

In all geological repositories, the long-term isolation of the waste from the biosphere relies 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:

1. The favourable 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?

Some examples of this are discussed in the following text. A more comprehensive discussion focused on the specific requirements for bentonite in several disposal concepts in clay rock and crystalline rock is given in D1.1

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. Swelling clay, therefore, stands out as a viable buffer material that meets all these requirements and therefore bentonite has 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 (Fig. 2.1). 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.

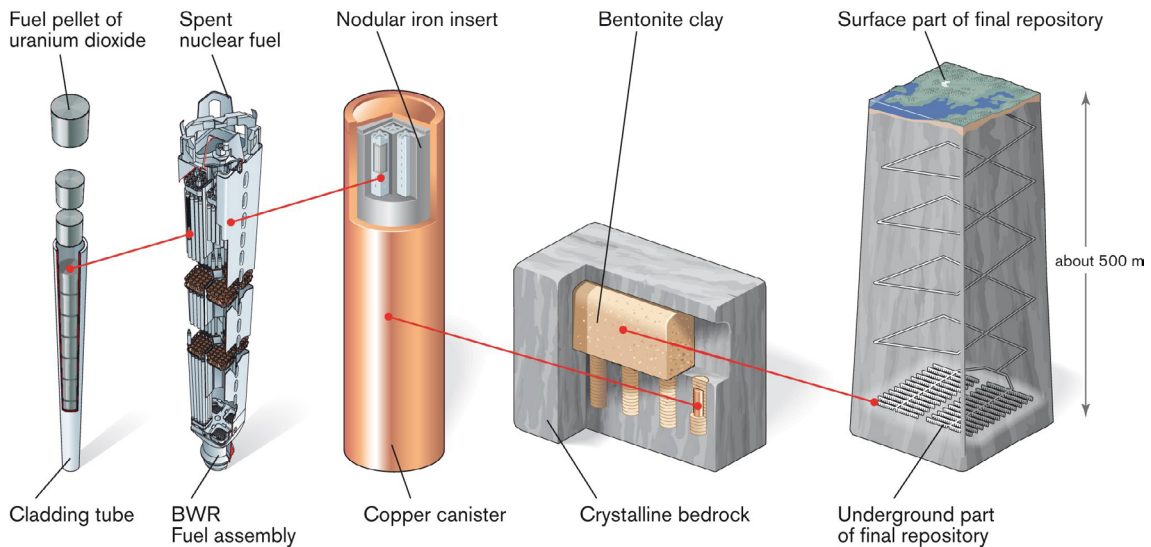


Fig. 2.1: The KBS-3 method

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).

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 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 the 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 concept and the boundary conditions, a hydraulic conductivity of 10^{-11} to $10^{-12} \text{ m s}^{-1}$ 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 water-saturated. 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 100 kPa to 1 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, stray materials in the repository, cement additives (such as super plasticizers), 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 is dependent 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 2,050 kg/m³ (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 Transformation (Requirement on Temperature)

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 100 °C is set in order to limit chemical alteration, while in clay rock environments, higher temperatures can be tolerated/justified, 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 is dependent on the properties of these barriers. 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 has been 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 ~ -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, as pyrite, 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 fulfils two basic mineralogical criteria: firstly, the montmorillonite content must be sufficiently high to provide and maintain the minimum swelling pressure, to ensure sufficiently low hydraulic conductivity, and to ensure the necessary range of stiffness and shear strength for the specified density interval; and, 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 $\sim 1,450 - 1,650 \text{ kg/m}^3$ for a fully saturated and homogenized buffer in the deposition hole/ or tunnel.

3 Outline of Cases Dealing with Identified Uncertainties

The four defined cases related to early evolution of the EBS that were defined early in the project to serve as a basis for integrating the project results are presented in Tab. 1.

Tab. 1: Cases related to early evolution of the EBS to be used as a basis for integration of project findings

Case number	Case description (principal focus)	Origin of the case (from WP1)	PEBS activities feeding into case assessment
Case 1	Uncertainty in water uptake in buffer (T <100 °C)	Discrepancies between standard THM model and FEBEX observations	<ol style="list-style-type: none"> 1. FEBEX mock-up data and modelling 2. THM Column Tests at Ciemat 3. EB experiment 4. Extension modelling of FEBEX in situ test
Case 2	Uncertainty in T evolution in buffer (T >100 °C) and impact on buffer properties	Lack of validation of TH model for high temperature and low saturation rate	<ol style="list-style-type: none"> 1. HE-E experiment, column experiments and modelling
Case 3	Uncertainty in HM evolution of buffer	Limited large-scale experiments	<ol style="list-style-type: none"> 1. EB experiment and modelling 2. HE-E experiment, column experiments and modelling 3. Febex mock-up and in situ 4. Stress-strain behaviour studies 5. Analytical model of homogenisation
Case 4	Uncertainties in geochemical evolution	Experiments vs. models of corrosion product/bentonite and cement/bentonite interactions	<ol style="list-style-type: none"> 1. GAME experiments and modelling 2. Interface studies (WP2.3) 3. Modelling in WP3.4

The second column represents only a broad descriptive name with a principal focus and it is not intended that the scope is limited only to the case description. In fact the assessment of barrier performance covers a broader range than indicated and there is inevitable overlap between the cases. The origins of the cases are highlighted in the third column. Case 1 originates with observations from the large-scale FEBEX experiment, designed as a study of the performance of the Spanish EBS concept, for which discrepancies have been observed between the time-dependent saturation behaviour of the bentonite and thermo-hydraulic model predictions. Case 2 originates with the Nagra's EBS concept, which foresees bentonite barrier temperatures well in

excess of 100 °C. Prior to the PEBS project, there were no large-scale experiments that permitted comparison between models and data for the thermo-hydraulic behaviour of such a system, so the HE-E experiment at Mont Terri provided an opportunity for such a test. Case 3 originates with the SKB EBS concept. It focusses on the hydro-mechanical performance of emplaced EBS material with different initial properties and how the properties of the barrier as a whole evolve over time, thereby significantly enlarging the knowledge that existed prior to PEBS. Case 4 deals with the results of experiments involving thermo-hydro-chemical interactions, in particular at internal and external interfaces of the bentonite barrier and comparison of results with geochemical models. The fourth column in Tab. 1 highlights the main experiments that are part of the PEBS project that contribute to improving understanding of the processes. It should also be noted that studies from outside of PEBS are also considered in the assessment of process understanding.

4 Progress in Detailed Process Understanding

4.1 Case 1: Water Uptake in the Bentonite Buffer ($T < 100\text{ }^{\circ}\text{C}$)

4.1.1 Introduction to the Process Understanding

The EBS of the repositories will be designed to operate in the long-term under saturated conditions while the bentonite will be initially placed unsaturated. Then, in the early stage of a repository, the bentonite will take up water from the surrounding bedrock (crystalline or clayey) becoming progressively saturated. The time scale for this process is strongly dependent not only on the bentonite barrier characteristics, but also on the hydraulic properties of the specific bedrock in each repository. The phenomena can be understood and their relevant features and consequences quantified using the numerical codes now available.

Several properties of the bentonite (and then safety functions) are directly related to its saturation degree. The most relevant ones are the following:

- Swelling capacity: development of the required swelling strain (to fully seal the initial voids and gaps in the barrier) and swelling pressure (specific mechanical requirements in the different performance assessment studies and minimization of the microbial activity).
- Hydraulic conductivity: low enough, in order to limit advective transport (including potential corroding agents towards the canisters) and provide a diffusion-controlled transport condition after saturation is achieved.
- Thermal conductivity: high enough to limit the possibility that excessive temperatures could develop in the bentonite.

To better assess the impact of the bentonite saturation process (and its time evolution) on the safety functions of the barrier, first it is worth noting that full saturation is not strictly speaking required to assure that the values of the safety function indicators (e.g., the hydraulic conductivity or the swelling pressure) are in the required ranges or beyond the defined conservative limits. In particular, it is proposed to consider the following two main phases in the bentonite saturation process:

- **Phase 1** (quasi-saturation): from the initial degree of saturation (S_r) during the barrier emplacement up to a high enough average value of S_r (above approximately 90 %, although this value can be different for each specific barrier design).
- **Phase 2** (full saturation): from the quasi-saturation state up to the full saturation, when the water uptake will stop completely (negligible bentonite suction).

As is shown in more detail in the following sections, to consider the proposed two phases improves the understanding of the potential relevance of some uncertainties related to the saturation process that have been recognized in some experiments (both in laboratory and "in situ") and in the available modelling. It can be stated that uncertainties and modelling discrepancies are mainly identified only in Phase 2, after the quasi-saturation of the bentonite, at a time when the barrier already fulfils its safety functions.

4.1.2 Contributing Issues from Experiments and Experiment Modelling in PEBS

4.1.2.1 Laboratory Tests

Laboratory tests have shown that, if water availability from the boundaries is sufficient, quasi-saturation of the bentonite is relatively fast (Phase 1 of saturation). Specifically, as part of the PEBS project, a series of infiltration tests have been performed under isochoric and isothermal conditions, and also applying low injection pressures. Three of these tests were performed with the pellets mixture used in the EB "in situ" experiment (Villar 2012); three with the HE-E sand/bentonite mixture (S/B); and two with the HE-E MX-80 pellets (Villar 2013b). Also, previously, starting in the framework of the FEBEX project, two other long-term column tests were performed (and dismantled after almost 12 years of operation), one (GT40) under thermal gradient and the other (I40) at isothermal conditions (Villar & Gómez-Espina 2009).

In these tests, most of the water intake took place in a short period of time. In contrast, the subsequent water intake (Phase 2 of saturation) was very small and slow. The contrast between the two saturation phases is more acute in low-density bentonite, in which Phase I is shorter and coincides with a sharper development of swelling pressure. More specifically, on the bentonite pellets mixtures tested the swelling pressure sharply increased at the first stage of hydration, then stabilised, and afterwards increased again, reaching practically an almost-equilibrium value when most of the water had already been taken up. Previous investigations had also showed that this is the usual pattern when saturating bentonite under isochoric conditions (Imbert & Villar 2006, Gens et al. 2011).

Upon saturation the pellet mixtures become a homogeneous material, with hydro-mechanical properties in the range of those for the saturated powder material of the same dry density. However, the water content and dry density gradients along the bentonite can remain even after having reached the equilibrium swelling pressure (Villar 2013b). In the long term, the water content and dry densities could become more uniform, although this is not evident for parts of the barrier saturated quickly in which very high water contents and low dry densities are soon reached (Villar 2012), since this large deformation could be irreversible, as the EB in situ test also revealed. Also, the analyses of the microstructure of the pellets mixtures at the end of the tests showed that most of the porosity was of diameter less than 7 nm.

Since the thermo-hydro-mechanical properties of buffer materials depend on dry density, they would be modified according to the dry density gradients in the barrier (Villar & Lloret 2007). It must also be taken into account that the density gradients are linked to water content gradients, and that the latter also affect THM properties. These gradients are expected to diminish over time, but it is possible that some density non-uniformities will persist indefinitely.

Both the water intake and the water content and dry density variations throughout the barrier will be greatly affected by the thermal gradient, even for temperatures below 100 °C. In the proximity to the container the water content could be very low until the temperature has considerably decreased, whereas in parts of the barrier outside the influence of the containers, a high saturation will be reached sooner.

For low density buffer materials, such as those used in the EB and HE-E tests, it is not considered that the computation of the degree of saturation, and consequently water intake, could be affected by the changes in pore water density, since this is close to 1.0 g/cm³ due to the low average dry density of the bentonite. However, for dry densities higher than 1.4 g/cm³ and as the ratio adsorbed/free water increases, the average density of the pore water in smectite is

higher than 1 g/cm^3 , which affects the computation of the degree of saturation and the rate of water intake during the Phase 2 of the saturation. The long-term permeability tests performed in FEBEX and MX-80 bentonite support this previously reported observation (Jacinto et al. 2012, Villar 2002, Marcial 2003, Villar & Lloret 2007).

For the Phase 2 of saturation of the barrier, during which the trend to a very low hydration rate causes significant differences between model predictions and measurements, non-standard processes have been invoked as possible causes for these differences. Among them is the effect of "non-Darcian behaviour" (i.e. the existence of a threshold gradient). This has been studied using the long-term permeability tests performed during PEBS. Possible threshold values around 200 or 1400 for the FEBEX bentonite have been deduced, depending on the dry density and injection pressure (Villar & Gómez-Espina 2009, Villar 2013a). Also, the preliminary data indicate that the threshold hydraulic gradient for MX-80 could be between 360 and 1200.

4.1.2.2 FEBEX Mock-up Test

The FEBEX mock-up test, a demonstration experiment of a clay engineered barrier at almost full scale and under controlled boundary conditions (Fig. 4.1), was installed in 1997. The observed THM behaviour shows good homogeneity throughout the test and has revealed several features of interest, such as the underestimation of saturation times and the major implications of the thermal aspects (ENRESA 2006).

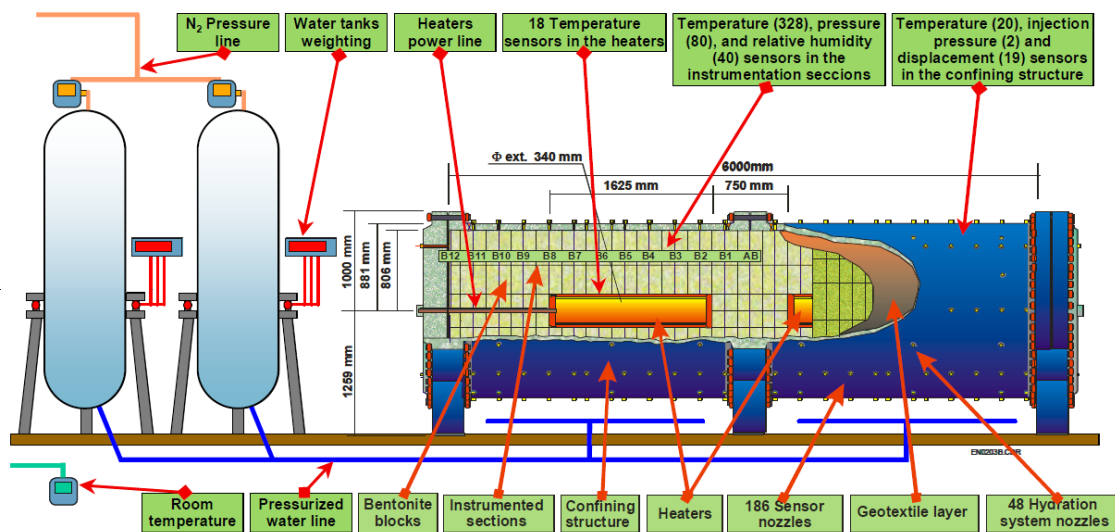


Fig. 4.1: FEBEX mock-up experiment at CIEMAT (Spain)

More than seventeen years after the start of the operational phase, it could be deduced from the most recent data available (Martin & Barcala 2014) that most of the barrier has achieved high degrees of saturation. In fact, after only three years the average value of the degree of saturation was higher than 90 %, according to the water intake measurements (Fig. 4.2).

Also, the relative humidity (RH) measurements indicate that even in the hottest part of the test, the external zone of the bentonite barrier ($\sim 0.25 \text{ m}$ had low suction values (between 10 - 15 MPa; and then was already near saturation) after only three years of the experiment life.

The saturation greatly affects the thermal conductivity. Values of thermal conductivity versus degree of saturation fit a sigmoidal curve (Villar 2002), with values from the dry state to the fully saturated one ranging from 0.6 to 1.3 W/m·K, and with a sharp increase between 40 % and 90 % of the degree of saturation.

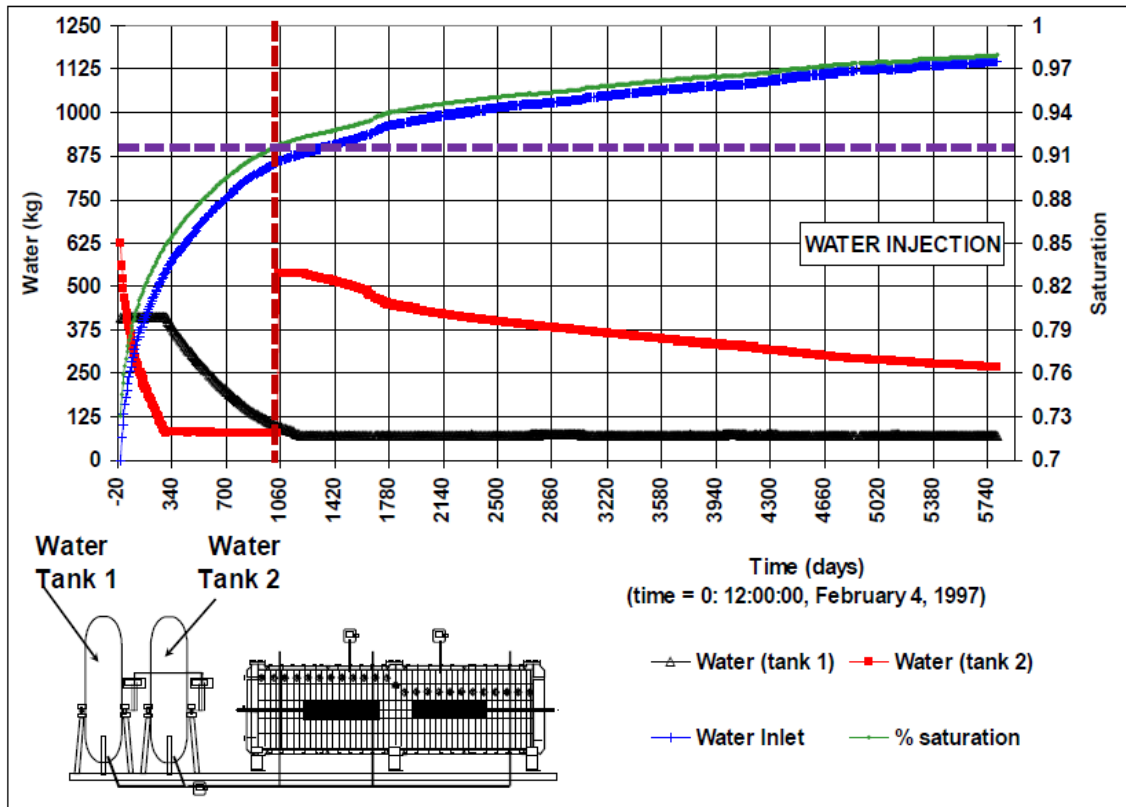


Fig. 4.2: Water intake and saturation degree in the FEBEX mock-up test

According to this, the estimated initial overall thermal conductivity in the barrier was around 0.8 W/m·K. After one year heating, when the drying of the internal material was most pronounced, the thermal conductivity in the inner bentonite close to the heaters decreased (probably down to 0.65 W/m·K), while it is estimated that the bentonite near the external hydration surface reached a value of 1.2 W/m·K (or even higher).

Current estimated saturation values indicate that most of the barrier has achieved thermal conductivity values higher than 1.2 W/m·K; except in the central zone close to the heaters, where thermal conductivity probably ranges between 0.8 to 1.0 W/m·K.

The main observations from this test and its modelling are the following:

- Temperature calculations produce good fits to data.
- The transient effects of evaporation (drying) and condensation (wetting) indicate the redistribution of water. Vapour migration through the barrier occurs even at medium-high degrees of saturation.

- Experimental measurements confirm that the water inflow (and water vapour outflow) is radial and homogeneous. The saturation of the bentonite is apparently controlled by the hydraulic properties of the bentonite and the local temperatures and/or thermal gradients imposed by heating, confirming significant differences between the "hot" and "cold" zones of the buffer as hydration progresses.
- The initial pressures depend on the specific location and installation of the sensor, the dry density of the block, and/or the development and concentration of local stresses. With time, values become homogeneous and stable as the buffer material saturates. Values for the different directions are converging to the swelling pressure values of the bentonite previously measured in the laboratory.
- The indicators of quasi-saturation are the following: high stable average values for swelling pressure and relative humidity, a positive trend in fluid pressure values, and very low water intake. All of them are occurring in most of the barrier, but not in the closest zone around the heaters. Although most the buffer is practically saturated, full saturation close to the heaters has not been reached yet.

From above, the following relevant conclusions may be drawn:

- The total saturation time is longer than foreseen by standard THM models.
- The buffer achieves the required performance even prior to full saturation, as shown by the sufficient and homogeneous swelling pressures developed and the homogeneous high thermal conductivity achieved.

The FEBEX mock-up test supports the evidence of the slowing down of hydration, under a specific thermal load and geometry. The long-term evolution of the test may provide a better understanding of the Phase 2 of the saturation, especially after its dismantling.

The conclusions above are derived from the homogeneous water supply conditions of the FEBEX mock-up test. Nonetheless, experience with the FEBEX in-situ test indicates that even in the case of water supply through a low transmissivity fracture, the saturation process of the bentonite proceeds rather uniformly (this might also be attributed to the fact that the permeability of the matrix was relatively high) (ENRESA 2006). The situation may be less clear in the case of a high transmissivity fracture in a very low permeable matrix.

4.1.2.3 EB "in situ" Experiment

The EB experiment was a full-scale demonstration test designed to represent the end of the transient phase (almost full saturation of the bentonite barrier and temperature approximately that of the host rock prior to repository construction). It was installed in a 6 m-long section of a gallery (2.7 m high and 3.0 m wide) excavated in the Opalinus Clay of Mont Terri (Fig. 4.3). A dummy canister (similar dimensions as the ENRESA and NAGRA reference canisters) was installed on the top of a bed of bentonite blocks and the remaining air volume ($\approx 28.4 \text{ m}^3$) of the section (finally sealed with a concrete plug) filled with a Granular Bentonite Material (GBM). An average dry density of 1.36 t/m^3 of the emplaced GBM was obtained. The experiment ran for more than ten years, under isothermal conditions and with artificial hydration during the first ~ 5 years. It was dismantled in the period October 2012 - January 2013 (Palacios et al. 2013), with more than 500 samples of the bentonite taken for on-site and laboratory analyses: dry density and moisture content; suction; pore size distribution; basal spacing; thermal conductivity; hydraulic conductivity; swelling strain and swelling pressure (Villar et al. 2014).

It should be noted that in the EB experiment there are inherent difficulties in emplacing the GBM because of interference due to the hydration tubes, thus the density achieved is lower than would be expected in a more ideal situation and the variation in density is probably significant. For example in the full-scale ESDRED emplacement study the emplacement dry density achieved was 1.44 - 1.51 t/m³ (ESDRED EU report).

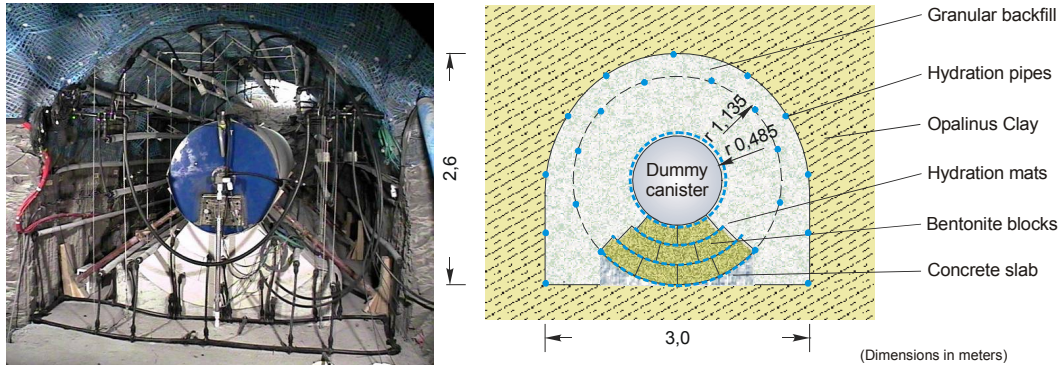


Fig. 4.3: EB in-situ experiment

According to the monitoring of the experiment during its more than 10-year life, the following main observations were made:

- Due to the artificial hydration system used, most of the bentonite saturated fairly quickly: in the first 1.5-year period, the water intake was equal to 15.2 m³ (while the estimated air volume before the water injection was 12.5 m³), although some initial water leakage was observed in the bottom of the concrete plug. Also, in this period, total pressures in the barrier as high as 1.7 MPa were recorded (while for a mean dry density of 1.36 t/m³ a maximum swelling pressure of only 1.3 MPa was expected). All the hygrometers (except one) showed full saturation.
- Following the relatively short first-period (1.5 years), during the remaining life of the experiment (~9 years) water intake was very slow (and no additional water was artificially injected after 5 years); and the recorded swelling pressures increased also very slowly, up to a recorded maximum value of 2.2 MPa.

After the experiment dismantling, the visual observations and the on-site and laboratory tests results have confirmed that the bentonite barrier was highly saturated and the bentonite blocks and GBM, which initially had significantly different dry densities, had developed rather similar average densities. More specifically, from the on-site density and moisture content determinations (Palacios et al. 2013), done at the experiment site and as soon as possible after taking the samples, the following can be concluded:

- The average degree of saturation is higher than 95 %. Moreover, during the dismantling operation it has been observed that, due to the time spent handling the samples (even if as short as possible), some drying and also some swelling (due to the non-confined conditions) of the bentonite could not be avoided before the tests were performed. The observed increase in the dimensions of the bentonite blocks placed under the canister indicates that they have swelled not only during the experiment's life but also along the dismantling operation. Then, it might be assessed that the actual degrees of saturation are even higher

than those calculated from the on-site test results. It has been estimated (Palacios et al. 2013) that very probably the actual degree of saturation varies from 98 % to 100 %.

- Although some degree of homogenization of the bentonite materials occurred (in the sense that the large initial density contrast between blocks and GBM has greatly diminished, the overall density range is still significant. There is also a clear trend for the moisture content to increase towards the bottom of the section (while the dry densities are lower than in the upper part) (see Section 4.3.2).

On the other hand, the results of the tests performed in the CIEMAT's laboratory have provided the following most relevant data of the characterization of the bentonite barrier dismantled:

- It has been confirmed that the bentonite was almost fully saturated. The obtained average degree of saturation is approximately 98 %.
- Most of the moisture content values determined range between 33 % and 43 %; and of the dry density between 1.24 t/m³ and 1.42 t/m³.
- One of the main objectives of the EB experiment was to determine if the hydraulic conductivity of the GBM (after saturation) is low enough, even if emplaced with relatively low average dry density (1.36 t/m³ in this case). The permeability tests performed on fifteen samples do confirm that, after saturation, the barrier had a low hydraulic conductivity (K_w): the values of K_w are equal to or lower than 5×10^{-12} m/s, except one (8×10^{-12} m/s), obtained with a very low dry density sample (1.18 t/m³), less representative of the overall barrier.
- Also, the measured thermal conductivity values are relatively high (as expected due to the saturation degree), from 0.90 to 1.35 W/m·K.
- Although the samples were almost fully saturated, they still had some suction (remaining capacity to absorb distilled water). Values ranging from 2.1 to 4.7 MPa have been measured. It is noted that also saturated samples can have suction (below air entry value). In that case, the suction is mainly related to the stress release rather than to desaturation.
- According to the pore size analyses, a relevant percentage of the porosity of the GBM samples was in the micro porosity range (diameter smaller than 7 nm), although a macro pore family (sizes between 3 and 35 μ m) has also appeared, which did not exist in the original GBM pellets. Also, macro porosity is predominant for samples with dry density lower than 1.30 t/m³.
- The dismantled bentonite still keeps some swelling potential: in the tests performed, swelling strain values as high as 22.5 % and swelling pressures up to 0.69 MPa have been measured.
- It is also noted that, after the additional water intake in the permeability and swelling tests, in general saturation degrees higher than 100 % (in some cases higher than 110 %) have been calculated, but assuming that the pore water density is 1.00 t/m³.

4.1.2.4 Insights from Modelling of THM Experiments

The current formulations and numerical models such as CODE_BRIGHT (Olivella et al. 1996) are capable of performing coupled thermo-hydro-mechanical analysis of a wide variety of geomaterials under saturated and unsaturated conditions. They are based on the simultaneous solution of a series of balance equations that express basic physical principles that have to be guaranteed at all times:

- Mass balance of water
- Mass balance of air
- Mass balance of solid phase
- Energy balance
- Momentum balance (equilibrium)

Within the framework of those balance equations, a number of phenomena are considered with the aim of taking into account all the relevant processes occurring in the engineered barrier as well as in the immediately adjacent rock. Those phenomena are captured by a series of constitutive laws and equilibrium restrictions that describe the various THM processes and their mutual interactions as well. The main ones are indicated below:

- Heat transport:
 - Heat conduction (Fourier's law)
 - Heat convection (liquid water flow)
 - Heat convection (water vapour flow)
 - Phase changes (Kelvin's or psychometric law)
- Water flow:
 - Liquid phase (Darcy's law)
 - Water vapour diffusion (Fick's law)
- Air flow:
 - Gas phase (Darcy's law)
 - Air solution in water (Henry's law)
 - Dissolved air diffusion (Fick's law)
- Mechanical behaviour:
 - Thermal expansion of materials (linear expansion law)
 - Behaviour of geomaterials dependent on stresses, suction, water pressure and temperature (elasto-plastic law)

Due to some differences between the computed results of the conventional THM modelling and observations (Fig. 4.4), involving especially the water intake of the barrier in the latter hydration phase, a number of modifications to the theoretical formulation and computer code have been incorporated, implemented and verified. The main ones are:

- Consideration of non-Darcy flow in the compacted bentonite, i.e. the possibility that there is: i) a threshold pressure gradient below which no flow takes place, and ii) a transition zone in which there is no proportionality between flow and pressure gradient. This requires the modification of Darcy's law in the description of the liquid flow (Sánchez et al. 2007).
- Incorporation of thermo-osmosis phenomena, i.e. the possibility that, in addition to water potential gradients, liquid flow is also driven by temperature gradients. This involves a change in the equation governing the water flow in the liquid phase through the addition of a flow term proportional to the gradient of temperature. Unfortunately, there is little experimental evidence for the determination of appropriate parameters for this new term.

- Consideration of the evolution of the microstructure of the bentonite in the engineered barrier throughout the hydration process. It has been experimentally observed that during hydration the pore structure of the compacted bentonite changes significantly due to the expansion of aggregates and the closure of the larger macro pores, thus changing drastically the value of intrinsic permeability of the material. This is taken into account by a modification of the elasto-plastic mechanical constitutive law that considers separately the microstructure and the macrostructure of the material and their mutual interaction (Sánchez et al. 2005, 2008, Gens et al. 2011).

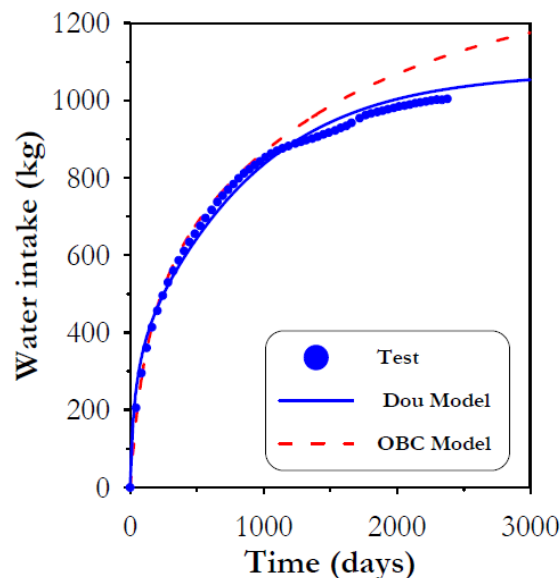


Fig. 4.4: FEBEX mock-up: water intake (Dou: Double porosity model; OBC: Operational base case model)

The numerical formulation and computer code has been applied to the infiltration tests carried out in the CIEMAT laboratory (Sánchez & Gens 2014). In general, the early stages of the tests are satisfactorily reproduced by the conventional formulation and models, indicating that the most relevant THM phenomena have been identified and correctly incorporated into the numerical models used. However, discrepancies between computations and observations are often noted concerning the latter part of the hydration phase. Naturally, those discrepancies can only be perceived in long-term tests. In this context, two long-term column tests on FEBEX bentonite, performed under a thermal gradient (GT40) and under isothermal conditions (I40), are of special interest in this context (Villar & Gomez Espina 2009). It is found that the very slow hydration observed in test GT40 cannot be reproduced using the conventional formulation. Indeed, even with the enhanced formulation, a satisfactory modelling is not quite achieved. There is the possibility that some vapour escaping from the apparatus can contribute to those difficulties; this issue will be clarified when the results from the dismantling become available. It should also be noted that slow hydration in the latter stages of the test have been observed in other thermal gradient tests performed in other laboratories and with different materials, see for instance the tests reported by Gatabin & Millaud (2005) on MX-80 bentonite. Indeed, Thomas et al. (2003) also reported unexpected late slow hydration in an isothermal large scale field test (ITT) performed in the AECL underground laboratory.

A large part of the computational effort has been devoted to the modelling of the FEBEX mock-up test. Because of its size, design and good performance, this test offers probably the most reliable and consistent set of long-term THM experimental results available at the present time. It therefore provides a good testing ground for the performance of the conventional and enhanced formulation for both the early and long-term stages of hydration. The analyses have been run under the following hypotheses (Sánchez & Gens 2014):

- Conventional formulation
- Enhanced formulation with the assumption of non-Darcy flow
- Enhanced formulation with the assumption of thermo-osmosis
- Enhanced formulation taking into account the evolution of microstructure
- Enhanced formulation combining the evolution of microstructure and thermo-osmosis

In all cases, the analyses have been taken to quite long times, of the order of 30 years. It has been found that the conventional formulation reproduces satisfactorily the early stages of the test but it fails to model the slow rate of hydration that occurs at later times. Discrepancies start to show after about three years, when the average degree of saturation of the barrier is about 90 %.

The three hypotheses (and the combined one) used in the enhanced formulation are able to model quite accurately the hydration of the mock-up test at all stages. Although the PEBS project is not designed to establish experimentally the potential existence and effects of those three phenomena, the performance of THM modelling of the FEBEX mock-up test (16 years of observations) gives some useful information on the effects likely to be associated with each one of the individual hypotheses considered.

Numerical modelling has shown that each of these possibilities is capable of providing results in quite good agreement with observations. However, numerical analysis, on its own, is unable to identify with certainty what is the phenomenon (or combination of phenomena) underlying the observed slowing down of hydration. However, there are noticeable differences between the predictions of the different hypotheses for the long-term situation (30 years). Therefore, availability of long-term observations of the mock-up test may help in identifying the relevant phenomena at the later phase of hydration.

4.1.3 Key Identified Uncertainties Related to Case 1

Related to the water uptake in the bentonite barrier, during PEBS no relevant uncertainties for the long-term safety functions have been identified. The uncertainties and modelling discrepancies are only recognized in the Phase 2 of saturation, and can be summarized as follows:

- Predicted hydration rates in that phase are larger than the experimental values; and then the actual time required to reach full saturation is longer than estimated by the models.
- The water volume taken up by the bentonite is larger than anticipated.

Nevertheless, the available models do provide robust guidance to assess and predict sufficiently well the Phase 1 (quasi-saturation) process, when the barrier already fulfils its safety functions. It is now possible to state that conservative values of the safety indicators (such as swelling capacity; hydraulic conductivity; and thermal conductivity) related to the bentonite saturation can be predicted with reliability. The context, from a long-term safety perspective, has been

clearly improved; and in any case, the residual uncertainty in water uptake during the Phase 2 might even be qualified as conservative: it is likely that advective flow conditions from the host rock towards the buffer will be maintained longer than estimated by the models.

4.1.4 Remaining Open Issues

From a scientific perspective, the final hydration process (Phase 2 of saturation) is not yet fully understood. Three hypotheses (see previous section 4.1.2.4) have been examined to explain the existing discrepancy between model predictions and experimental data. Numerical modelling has shown that each of these possibilities is capable of providing results in approximate agreement with observations but, on their own, they are unable to identify with certainty what is the phenomenon (or combination of phenomena) underlying the observed slowing down of hydration.

Further research developments about this subject should improve the in-depth knowledge about the evolution of the microstructure of the bentonite, the properties of the adsorbed interlayered water, and about the swelling pressure development.

4.2 Case 2: Uncertainty in T Evolution in Buffer ($T > 100$ °C) and Impact on Buffer Properties

4.2.1 Introduction to the Overall Process Understanding

Earlier work assessing the performance of the bentonite barrier beyond 100 °C is reviewed in Wersin et al. (2007). It was concluded that relevant data for repository conditions from both laboratory and field studies is scarce, but nevertheless yields a fairly consistent picture. No significant changes of hydraulic and mechanical properties have been reported for bentonite materials exposed to temperatures of at least 120 °C under wet conditions. The data suggest significant cementation and perhaps also illitisation effects occur at 150 °C and beyond. Interestingly, natural analogue bentonite samples that showed substantial cementation and illitisation effects still displayed rather favourable hydraulic properties. Under dry conditions, bentonite is stable to higher temperatures maybe as high as 350 °C (Wersin et al. 2007).

A more recent review of the scientific understanding of the processes related to bentonite used as an engineered barrier is given in Sellin & Leupin (2013), which focussed on bentonite in general at repository relevant temperatures. In their review, the current understanding regarding processes related to the early re-saturation phase were described, including heat transport, water uptake, evolution of the swelling pressures, diffusive transport, gas dissolution and transport, bentonite interaction with metal corrosion products, bentonite interaction with cementitious materials, microbial effects and thermal alteration of bentonite. Based on this review, the authors identified the following issues related to Case 2 to be safety relevant and in need of further work:

- Effects on bentonite of long periods of unsaturated conditions with elevated temperatures, including the stability of bentonite under non-isothermal conditions and different degrees of saturation
- Redistribution of mass during swelling, and its different impact on pellets and blocks when applied in the same setup
- Redox stability of bentonite under highly reducing conditions (induced by the presence of hydrogen gas)

While the first two issues are addressed by the PEBS project (either in the laboratory, URL or as part of review work) the latter is clearly beyond the scope of the project.

With the HE-E experiment (Fig. 4.5) a 1:2 scale version of the Nagra emplacement concept in two symmetrical sections, designed and constructed as part of PEBS, substantial progress has been made in the understanding of the behaviour of a combination of bentonite blocks and granular material (bentonite and sand/bentonite) exposed to temperatures up to 140 °C at the URL scale. The monitoring data and the modelling thereof allowed identification of remaining uncertainties, although future monitoring of the HE-E will allow refining this further. The experiment was supported by laboratory column tests allowing characterisation of the granular materials individually under similar conditions.

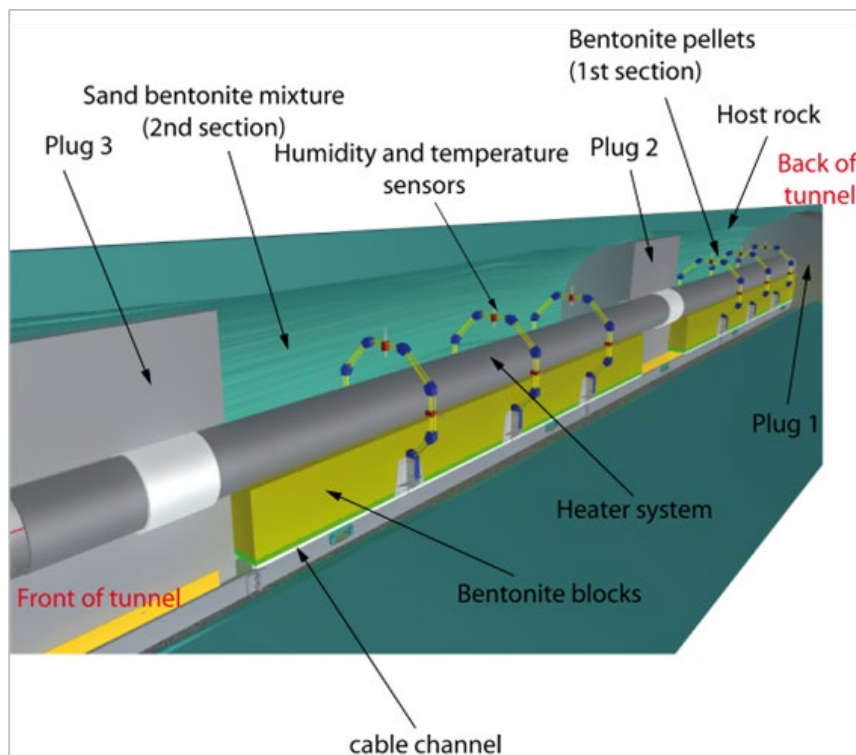


Fig. 4.5: Schematic layout of the HE-E experiment at the Mont Terri URL showing the section in the back of the tunnel filled with bentonite pellets and the section in the front of the tunnel filled with sand/bentonite

Within PEBS it was confirmed that while the characterisation of bentonite performance below 100 °C is largely established, the information regarding bentonite at temperatures >100 °C is less abundant. This is especially the case for granular bentonite material (GBM; manufactured bentonite as grains or pellets) although recently retention behaviour of this material was characterized at two different temperatures, however, below 100 °C (21 °C and 80 °C; Rizzi et al. (2012)). As part of the FE experiment, a full-scale simulation of the Nagra emplacement concept (in construction as of February 2014; Müller et al. 2014) further detailed characterisation of granular material at elevated temperatures will take place.

The contributions of the various aspects of PEBS to the increased understanding are discussed in the following paragraphs and followed by a discussion related to the remaining uncertainties

Although the focus is on safety-relevant uncertainties, no link with the safety functions is established here. This will be realised in paragraph Chapter 5. In this chapter the focus is on framing the uncertainty in process understanding either by describing and assessing the conceptual uncertainties or by providing a range for parameter uncertainties where possible.

4.2.2 Contributing Issues from Experiments and Experiment Modelling in PEBS

4.2.2.1 Laboratory Testing of Properties above 100 °C

Within the PEBS project laboratory work focussed on two main areas. One area of focus were column tests performed with the granular bentonite material and the sand/bentonite mixtures of the HE-E experiment to characterise their thermo-hydraulic behaviour when subjected to temperatures similar to the HE-E experiment. The second area of focus was on stress-strain properties of bentonite specimens exposed to increased temperatures (up to 150 °C).

In the column tests cells of granular bentonite material (referred to in the following discussion as B), and sand/bentonite (S/B) mixture are hydrated through the upper surface whereas the lower surface is heated at a constant temperature. The infiltration tests for the HE-E are being performed in cylindrical cells similar to the cells used during the FEBEX and NF-PRO projects (Villar et al. 2005, 2008). The nominal internal diameter of each cell is 7 cm and the inner length 50 cm. The bodies of the cells were made of Teflon to prevent as much as possible lateral heat conduction. The bottom part of the cells has a planar stainless steel heater. Inside the upper steel plug of the cells there is a small reservoir in which water circulates at room temperature. In this way, a constant temperature gradient between the top and bottom of the sample is imposed. Hydration with Pearson synthetic water takes place through the upper lid of the cell. The different components of the system were described in detail in Villar et al. (2012). The columns were manufactured by filling the cells in seven 7-cm high layers. The material was simply poured into the cell. The resultant dry density was 1.45 g/cm³ in the case of cell S/B (w = 4 %) and 1.53 g/cm³ in the case of cell B (w = 6 %).

The water volume intake, the heater power, the axial pressure on top (in cell B), and the relative humidity (RH) and temperature (T) at three different levels inside the clay are being measured as a function of time (Villar et al. 2014). The tests started with two heating phases, with the heaters set consecutively at 100 °C and 140 °C. Once the relative humidity recorded by the sensors remained stable, hydration started at a heater temperature of 140 °C. Since the water availability at the Mont Terri gallery is very limited, only a small pressure, given by an equivalent 60-cm high water column, was applied to the saturation water. The hydraulic boundary condition is still quite different from the hydraulic boundary condition in the HE-E experiment where the water supply to the EBS materials is determined by the hydraulic conditions in the Opalinus Clay driven leading to extreme low inflow. Furthermore, due to the different geometry and thermal conductivity (due to the differences in water content) of the materials involved, the temperatures in the column tests are lower than those in situ. This needs to be kept in mind when comparing the observations from the column experiments and the HE-E experiment.

The heating phase of both tests showed that the thermal conductivity of the as-emplaced B material is low, which caused a large difference in temperature between the heater surface and the sensor located at 10 cm, generating a high thermal gradient near the heater, and low temperatures in the rest of the columns. The stabilisation of the temperature in these materials was very quick.

The movement of water in the vapour phase as a result of the thermal gradient was indicated by the sharp increase of relative humidity recorded by the sensors closest to the heater -followed by a continuous decrease- and the slower increase recorded by the other two sensors. The different permeability of both materials was made clear in the different pace and extent of this water redistribution process in the vapour phase. Thus, the initial increase of relative humidity at 10 cm from the heater and in the upper part of the column was faster in cell S/B. Also the relative humidity gradient at the end of the heating phase was sharper in cell S/B than in cell B, due to the lower permeability and higher water retention capacity of the bentonite pellets, which meant that before hydration, the highest relative humidity in cell B was recorded in the middle of the column.

The lower permeability of the pellets was again highlighted by the fact that after more than 300 h of hydration, the upper sensor in cell B had not yet recorded any RH change, while by this time the lower sensor in cell S/B had already recorded the arrival of the hydration front.

Fig. 4.6 shows the current temperatures and relative humidities for the two tests (February 2014). Although, as explained above, during the heating phase the temperatures measured were higher in the B cell, this reversed during hydration. The reason is probably the increase in thermal conductivity of the S/B material after being completely saturated. The hydraulic state of both materials is totally different, the S/B mixture having reached full saturation (two of the sensors are flooded) while the B column shows still a steep relative humidity gradient, with approximately the 15 cm closest to the heater having a relative humidity below the initial one.

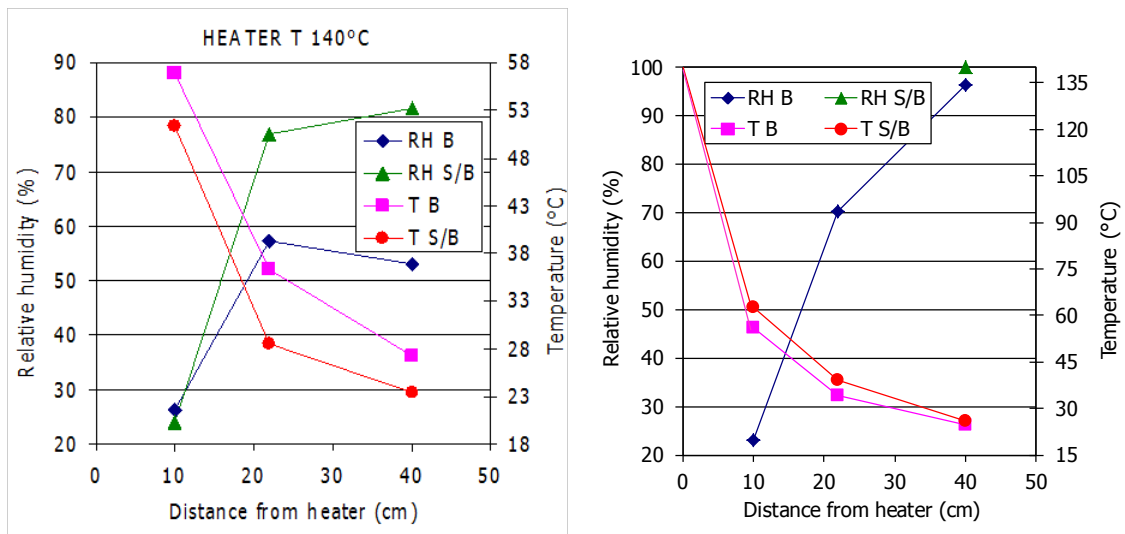


Fig. 4.6: T and RH along the buffer columns (heater at 140 °C) just before saturation (left) and after 682 days hydration (cell S/B) and 620 days hydration (cell B) (right) (Villar et al. 2014)

The axial pressure measured during the hydration phase seems to have stabilised at a value of 1.4 MPa, which is far from the equilibrium swelling pressure of MX-80 bentonite compacted at dry density 1.53 g/cm³. This is not surprising since the bentonite has still a low degree of water saturation and materials with double porosity (macro/micro) are known to display a non-monotonic development of swelling pressure (Imbert & Villar 2006, Gens et al. 2011). No swelling is expected for the S/B mixture.

The impact on the materials at hand can only be evaluated through post-mortem analyses, which has not been completed as part of the PEBS project. However, earlier similar tests were completed and the outcomes are interpreted jointly in the following paragraphs.

Two thermo-hydraulic tests were performed in an experimental setup similar to that described above; using MX-80 powder bentonite compacted at a dry density of 1.69 g/cm^3 with a water content of 17 % (Gómez-Espina & Villar 2010a, b, 2013). The column length was 20 cm, the heater was set at $140 \text{ }^\circ\text{C}$ and saturation with deionised water took place through the top surface of the columns. The tests reproduced the large-scale Temperature Buffer Test (TBT; Sandén et al. (2007)) performed at the Äspö HRL and their durations were 500 and 1500 days. In these experiments deionised water was injected on top of the cells at a pressure of 0.01 MPa.

The steady-state temperatures inside the bentonite were $82 \text{ }^\circ\text{C}$ at 6 cm from the heater and $40 \text{ }^\circ\text{C}$ at 16 cm from the heater. The long time needed to reach hydraulic equilibrium during the initial heating phase, showed the low permeability to water vapour of the bentonite with a degree of saturation of 75 %. At the end of the tests sharp water content gradients were observed along the columns, as well as inverse dry density gradients. The water content in the 5 cm closest to the heater was much lower than the initial value, and the bentonite had shrunk in this area. This indicates that the bentonite dry density had increased with respect to the initial dry density of the blocks. However, in the hydrated parts of the bentonite column the dry density decreased well below the initial value. Hydration modified the bentonite microstructure, in particular the pore size distribution, the smectite basal spacing and the specific surface area. In addition, an overall decrease of the smectite content with respect to the initial value took place, especially in the most hydrated areas - where the percentage of interstratified illite increased- in the longest test. On the other hand, the content of cristobalite, feldspars and calcite increased. Smectite dissolution processes (probably colloidal) occurred, particularly in the more hydrated areas and in the longest test. Due to the dissolution of low-solubility species and to the loss of exchangeable positions in the smectite, the content of soluble salts in the pore water increased with respect to the original one, especially in the longer test. The solubilized ions were transported; sodium, calcium, magnesium and sulphate having a similar mobility, which was in turn, lower than that of potassium and chloride. The cation exchange complex was also modified. A decrease in the exchangeable sodium content closest to the heater was observed, whereas the exchangeable calcium content increased. Calcite precipitated in the hottest areas. The pore water evolved from Na-HCO_3^- to Na-SO_4^{2-} in the hydrated areas and Na-Cl in the drier areas.

The following conclusions can be drawn regarding the column experiments. A steady thermal gradient is quickly established in bentonite-based buffers, irrespective of their dry density and water content. Consequently, the temperatures inside the buffer barely change during the heating and hydration phases and are just related to the proximity to the heater. They are also greatly affected by the boundary conditions, and this is why they are not comparable in laboratory and in situ tests (Villar et al. 2012b). However, the permeability to water vapour is highly dependent on the degree of saturation, and the water content redistribution triggered by the thermal gradient will be slower the higher the degree of saturation. The combination of thermal and hydraulic gradients gives rise to water content and dry density gradients along the buffer which are very persistent. The tests in columns performed so far have not allowed checking their reversibility, because they have not run for long enough. The swelling capacity of MX-80 bentonite under repository conditions remained for temperatures up to at least $82 \text{ }^\circ\text{C}$.

The study on stress-strain properties of bentonite specimens exposed to increased temperature was reported in the Deliverable D2.2-12. The influence of increased temperature was quantified

by measurements of stresses and strains during the unconfined compression test and also by measurements of swelling pressure and hydraulic conductivity made after the heating.

In the test series with increased temperature the specimens were exposed to short-term heating in a laboratory oven during 24 h. Specimens of MX-80 (Na dominated Wyoming bentonite produced by American Colloid Co.) were exposed to 90 °C, 120 °C and 150 °C and specimens of FEBEX (Mg-Ca dominated Almeria bentonite produced by Minas de Gádor S.A. (now Süd-Chemie Espana)) were exposed to 90 °C. The heating was made both after and before full saturation where heating after full saturation meant heated with a controlled water pressure to a maximum of 600 kPa while heating before full saturation in this study meant heating at a high degree of saturation without control of the water pressure but under sealed conditions.

The following correlations were seen in the results from the study:

- A tendency of increased deviator stress at failure with increased temperature after short-term heating but significant deviations only after heating to 120 °C and 150 °C
- Significant decrease in strain at failure only after short-term heating to 150 °C
- Small decreases in swelling pressure and hydraulic conductivity after short-term heating

4.2.2.2 Outcomes of the HE-E Experiment

The main element contributing to the increased process understanding of bentonite barriers above 100 °C in the PEBS project was the HE-E experiment, a 1:2 scale heating experiment installed at the Mont Terri Rock Laboratory, which was designed and constructed as part of PEBS and initiated in June 2011 (Gaus et al. 2014).

The HE-E experiment is designed as a model validation experiment at the large scale and subjects the EBS and the host rock to temperatures higher than in situ experiments performed in the past, using realistic heating rates while undergoing natural hydration from the low permeability Opalinus Clay host rock.

At the end of the PEBS project, the HE-E had been operating for almost 32 months successfully. As of February 2014 the major outcomes can be summarized as a follows.

The observed temperature increases in EBS and Opalinus Clay are in line with those predicted by the design calculations (slight variations are attributed to differences in model setup and conceptualization). The EBS is characterized by a very strong temperature gradient owing to its low thermal conductivity in its very dry state especially in the inner part of the buffer. At the Opalinus Clay interface a temperature below 50 °C is registered. The heat pulse causes a further drying of the inner part of the buffer, whereby the initial water content is further reduced, below the water content at emplacement for both the granular material and the blocks. A complex development of the humidity profiles takes place which is strongly determined by the different water contents and densities of the materials at installation, high sensitivity to changing two-phase flow parameters and the impact of vapour diffusion in a changing porous matrix. The vapour is driven out, most likely in a radial pattern and part of the increase in relative humidity at the interface between the EBS and the host rock can be attributed to condensation of vapour rather than inflow. The highest temperatures (>100 °C) are thus prevailing in an EBS with a very low water content (below <20 %RH). This observation is of importance because under these conditions of very low water contents, chemical reactivity will be hampered by water availability. As long as high temperatures at the heater surface are maintained, these dry conditions are to prevail. The natural water inflow from the Opalinus Clay is occurring slowly.

Although pores become saturated fairly rapidly (more so in the sand/bentonite than in the bentonite), after 32 months only at distances in the Opalinus Clay >1 m from the tunnel wall a hydraulic pressure is registered. The hydraulic pressure front is progressing toward the EBS, but how long this will take and when an equilibrium state will be reached if keeping the 140 °C constant cannot be determined from the current dataset. This can only be assessed using modelling.

The measured temperatures and relative humidities in the blocks and the granular materials are dominated by the distance of the measurement point to the heater and not by the differences in material properties although conditions at emplacement were somewhat different. This rapid homogenisation can also (partly) be explained by vapour movement and is observed for the first time in the HE-E, as this is the first large scale, high temperature, experiment with different materials.

As predicted by the models, a hydraulic pressure increase, associated with the differential thermal expansion of the Opalinus Clay and the porewater, is observed in the saturated Opalinus Clay at a larger distance from the tunnel. The porewater pressure increase, which started developing shortly after switching on the heaters, developed further after the heater temperature was held stable at 140 °C and was maximal at about 3 m from the tunnel wall where an overpressure of 1 MPa developed, slowly flattening off a couple of months after the heater temperature stabilised. These overpressures were within the expected range, although it cannot be fully excluded that the overpressures are influenced by the presence of the tunnels nearby acting as a constant pressure boundary at 1 bar.

Based on the seismic transmission measurements, a variation of the derived P-wave velocity evolution was observed with pronounced dependencies on the orientation of the travel paths to the bedding (anisotropy) and their distances from the S/B-Opalinus Clay interface. A sequence of decreasing and increasing P-wave velocities points to the creation and sealing of invisible micro cracks within the first 50 cm from the S/B-Opalinus Clay interface, most probably caused by desaturation, the thermal pulse and the saturation and porewater pressures changes. Seismic methods have been shown to be a very sensitive tool for the continuous characterization of changes in rock properties although no clear link between the seismic parameters and the physical phenomena could be established.

4.2.2.3 Modelling of HE-E Experiment and the Column Tests

The results of the modelling are extensively discussed in D3.2-2. In this section only the aspects relevant for the behaviour of the barriers above 100 °C are derived.

The temperatures in the buffer materials and in the Opalinus Clay near field (<2 m) are reasonably well reproduced with the various model approaches. Modelled temperatures are highly sensitive to the relationship between thermal conductivity and water saturation especially for low water saturation. Detailed characterisation of this aspect of the materials at hand is recommended.

There is reasonable agreement between models and measured TH parameters in early re-saturation. The most advanced THM models are capable of reproducing the dry-out zone which is developing over almost half the radial thickness of the bentonite. This is less the case for the TH models.

An important effort has been devoted to the characterization of the thermo-hydraulic properties of the materials involved, in particular, buffer materials. A compilation of several laboratory

tests, site measurements and some additional back-analysis have been carried out for this purpose. However, some uncertainties regarding the water retention curve of the granular bentonite materials and the dependence of their thermal conductivity and water permeability on the degree of saturation still remain. The correlation between the parameters complicates the calibration process.

Models that differentiate in their setup between the initial state of the emplaced materials and their properties are capable of capturing the evolution of these materials at the start of the experiment. However, the homogenisation in temperatures and relative humidities across the material boundaries that develop after a certain time (approximately 1 year) cannot be reproduced in detail as the model results are determined by the initial material parameters.

The build-up of hydraulic pressures around the tunnels is slow and the area close to the tunnel surface is still in suction. It will require some years of monitoring to adequately test the models for capturing this behaviour.

The size and the trend of the hydraulic overpressures can be reasonably well reproduced with various models. To obtain satisfactory results the hydraulic conditions in the Opalinus Clay at a larger scale prior to the experiment initiation need to be established, which is not trivial in a URL as its presence causes an important disturbance of the hydraulic field compared to a natural (hydrostatic) regime.

Although, due to the complexity of a full-scale field experiment, it is always difficult to match all the measurements with the numerical outcomes, the numerical results are in good agreement with the measurements and it demonstrates the model capability of capturing the evolution of the heating experiment during its initial phase. The overall conclusion is that the tested TH(M) models have been shown to perform adequately for modelling this high temperature URL experiment, the performance of the barriers is well reproduced and the confidence in the longer term calculations, beyond the experimental timescale, is reinforced.

4.2.3 Key Issues Identified during PEBS Representing Uncertainties related to the Case 2

4.2.3.1 THM Properties (Swelling Pressure, Gas and Water Permeability, Plasticity, CEC) Evolution at $T > 100$ °C - Review of Existing Information

The systematic characterisation of THM properties above 100 °C has not been conducted, however certain trends towards higher temperatures can be observed. The swelling capacity of the FEBEX bentonite decreases slightly for temperatures close to 100 °C (Villar & Gómez-Espina 2009, Villar et al. 2010). As well, a decrease in swelling pressure as a function of temperature was observed (of the order of 30 %, a very broad indicative value based on extrapolated trends). Nevertheless the deformation of bentonite is more dependent on the stress applied than on the temperature. The water saturated permeability of the FEBEX bentonite increases with temperature, approximately as expected from the change in water viscosity. In addition, the water retention capacity of the bentonite (both FEBEX and MX-80) decreases clearly with temperature, especially when it is above 60 °C and when the density of the bentonite is high and the water content low (Villar & Gómez-Espina 2008).

It can be concluded that, in spite of these observations, the FEBEX bentonite remains suitable as a sealing material in HLW repositories (from the hydro-mechanical point of view) for

temperatures up to 80 °C, as it keeps its low permeability and self-healing ability. The extrapolation of results points out to the preservation of properties for at least up to 100 °C.

Overall, the observed effects of temperature on the hydro-mechanical properties (decrease of swelling pressure, increase of permeability, decrease of water retention capacity) can be qualitatively explained by considering the transfer of high-density interlayer water to the macro pores that is triggered by the increase in temperature. The different behaviour of smectites exchanged with monovalent versus divalent ions can be explained by taking into account the different interlayer/diffuse-double-layer water ratio in both.

Deviating test results, compared to reference tests, were seen on for example stresses and strains at failure or hydraulic conductivity determined on samples from large-scale field tests (Dueck et al. 2011, Karnland et al. 2009, Åkesson et al. 2012). It could not be excluded that the deviations were caused by the increased temperature.

4.2.3.2 Impact of Heating under Partially Saturated Conditions on Swelling Properties

Bentonite is invariably emplaced around SF or HLW canisters in a partially saturated state. Typical moisture content values for prefabricated blocks with a dry density of about 1,580 kg/m³ are in the range of 15 - 20 % (50 - 80 % of saturation), depending on requirements. In the case of granular bentonite backfill, typical moisture contents may be in the range of 4 - 6 %. As a consequence of the under saturation at emplacement and the heat generated by the disposal canister, some drying of bentonite will occur close to the canister. Inflow of water from the rock will gradually increase the moisture content of the bentonite and the saturation front will gradually move towards the canister. Results show that the homogeneity of the buffer during re-saturation is related to the degree of localized inflow. For example, in the FEBEX in situ experiment, concentric rings of relatively uniform moisture content are observed after 15 years, ranging from 100 % saturated near the rock to 80 % saturation near the heater (Lanyon & Gaus 2013). In contrast, strongly localized fracture inflow could cause variations in moisture content in the bentonite (e.g. as in the canister retrieval test in the Prototype Repository at Äspö). In the case of Opalinus Clay, the slow porous medium inflow appears to lead to circumferential rings with relatively homogeneous moisture distributions similar to the case FEBEX (Gaus et al. 2014).

The consequences of the slow inflow and thermal gradient are that the unsaturated bentonite close to the canister will be exposed to elevated temperatures for some decades. The question of how this may influence the subsequent swelling properties after full saturation is discussed here. It is noted that no new work was done in this area in the PEBS project; nonetheless, this topic has been the subject of many studies, including some performed parallel to the PEBS project, and the results are highly relevant to the question of safety-relevant properties during the period after the thermal and re-saturation transient.

Several studies of the effect of unsaturated conditions on bentonite swelling capacity were performed after the publication by Couture (1985) of results showing that MX-80 bentonite powder experiences a great reduction in expandability after treatment with steam at 150-250 °C. Oscarson & Dixon (1989) also found similar results for uncompacted Avonlea bentonite (80 % montmorillonite) down to temperatures as low as 110 °C and noted that the material would reswell when ultrasonically treated and metaphosphate was added. Results of a more comprehensive series of experiments of the type performed by Couture are discussed in Birgersson et al. (2014). Large differences in expanded volume between reference and vapour exposed material were found in some bentonites when bentonites were actively dispersed prior

to testing. The results were qualitatively similar to what previously has been reported by Couture. However, the findings concern the ability to form much expanded gels, and the short-term exposure to water vapour at the temperatures 150 and 200 °C did not significantly change the swelling capacity.

Oscarson and Dixon (1990) also performed studies on bentonite compacted to dry densities of 1.0 - 1.3 Mg/m³. Samples were compacted with water to give initial saturation levels of 0, 50 and 85 %, and a 100 % saturation level sample was achieved by saturation after compaction. Swelling pressures and hydraulic conductivities were measured before and after heating the samples at temperatures of 90, 125 and 150 °C. No significant changes in swelling were observed with the possible exception of one sample at 150 °C, nor did hydraulic conductivity change significantly. None of these studies found any differences in montmorillonite content after thermal treatment, thus the results have been attributed to aggregation and/or cementation phenomena.

Further work was done by Haas et al. (1999) and Madsen (1998), who passed steam through compacted samples with dry densities of 1.4 and 1.75 Mg/m³ at 150 °C. For a density of 1.4 Mg/m³, swelling pressure declined from 3.8 MPa to 2.2 MPa after treatment at 110 °C and from 3.6 to 1.9 MPa after treatment at 150 °C. Repeated cycles resulted in an asymptotic value of about 2 MPa. Again no mineralogical changes were observed. It is not clear that the experimental method, which involved passing a large amount of water as steam through the samples, is a meaningful simulation of the condition in a repository.

Pelletized granular bentonite was studied by Pusch et al. (2003), who found dense bentonite pellet samples had little reduction of swelling pressure after treatment at 110 and 125 °C and a reduction to about 50 % of the value for pellets of the same density at 150 °C upon exposure to water vapour. Valter and Plötze (2013) observed in a series of experiments that some reduction in swelling under saturated conditions occurred, largely in the case of samples heated to above 120 °C.

All the studies note that there is no detectable mineralogical change in thermally treated material, instead the property changes seem to be related to physical aggregation and possibly some silica cementation. Material is observed to re-swelling only when sonicated, usually also in the presence of phosphate. The studies include short-term experiments (as little as a few days) and longer-term experiment (more than six months). No appreciable differences in property changes appear to exist in relation to the different durations of treatment.

Regarding mineralogical transformations under unsaturated conditions, smectite-to-illite transformation rates are considerably slowed depending on the saturation state. This was shown by the experimental study of Whitney (1990) indicating retardation of illitisation in unsaturated conditions. This result was attributed to the restricted transport of solutes.

The results discussed above are not completely consistent, but nonetheless show that significant reduction in swelling pressure can occur in some cases above 110 - 120 °C, although in most cases in which swelling pressure was measured, values of more than 50 % of initial values were observed. The reasons for inconsistency of results may relate to hysteresis factors and also to rapid cooling of samples to room temperature in experiments prior to measurement of swelling pressures. Because re-saturation would be expected to occur in a repository while the bentonite is still at elevated temperatures, it is not clear that the experiments adequately simulate the evolution of repository conditions. It is noted that if fully saturated samples are exposed to short-term laboratory heating as high as 150 °C and swelling pressure and hydraulic conductivity are measured after cool-down, property changes are small compared to reference

untreated samples (Dueck 2014). Furthermore no significant changes were found after the six-year duration LOT study, performed at Äspö, in which the bentonite experienced temperatures up to 130 °C (Karlund et al. 2009).

4.2.3.3 Calibration of Parameters at the URL Scale and Use in the THM Models

Because of the limited characterisation of the barrier materials above 100 °C mentioned above, it has proven advantageous to calibrate the key parameters of the THM formulation at URL scale using the observations gathered in the performance of the HE-E test (1:2 scale). As the most important driver of the transient processes is the evolution of temperature, the most important calibrated parameter has been the thermal conductivity of the barrier materials and, especially, their dependence on degree of saturation. By examining the distribution of temperatures across the barrier, it has been possible to derive a realistic variation of thermal conductivity with degree of saturation. As heat transport is nearly exclusively due to conduction, this automatically leads to a good prediction of temperatures and their evolution. The second key parameter to be derived at URL scale is the unsaturated permeability. This has been calibrated from the evolution of relative humidity across the barrier. In this case the modification of the initial assumed value has been small. These calibrations have been confirmed and partially validated using the non-isothermal column tests performed at CIEMAT, although the distributions of temperatures in those tests are significantly different.

Observation of thermally-induced pore water pressures in the rock contribute also to the calibration of the stiffness and permeability of the rock. Such pore water pressure increases are the result of the interplay of pore water pressure generation (caused by the differential thermal expansion of water and solid phase and modified by the rock skeleton deformation) and pore pressure dissipation (controlled by the permeability of the rock). However, the testing time required for reliable calibration of rock parameters is longer than for the engineered barrier and has not been fully achieved within the lifetime of the project.

As a result of these calibrations at large scale, critical TH model parameters acquire a much higher degree of confidence. No calibration has been possible for mechanical parameters of the engineered barrier as there are no observations of mechanical variables and the degree of back-coupling from the mechanical component to the thermo-hydraulic one is small.

4.2.3.4 Adequacy of Using Existing THM Models above 100 °C

The THM formulation developed and validated for temperatures below 100 °C can be extended without modifications to temperatures above that value. As discussed in Olivella & Gens (2000), from a physical point of view, there is nothing remarkable associated with the 100 °C temperature. The water present in the bentonite (porous medium) will not boil at that temperature; the transformation from liquid water to vapour will be an ongoing process in which phase change depends on the matric potential of a particular unit of water. As the matric potential varies gradually with the degree of saturation, liquid water will become vapour also gradually without requiring any changes in the basic physical laws used in the THM formulation. Naturally, there are some variations of retention curve, water permeability and surface tension with temperature but those variations can be readily taken into account without any modification of the basic formulation.

This approach has proved successful, for instance, in the modelling of laboratory tests performed on MX-80 bentonite by Gatabin & Billaud (2005) where a temperature of 150 °C

was reached. A good reproduction of the main results was achieved without altering the basic THM formulation. Further evidence is provided by the successful modelling in this project of the early stages of the HE-E experiment.

An important observation, however, is that at those high temperatures, the assumption, often made, that gas pressure is atmospheric and constant can no longer be adopted. As the vapour pressure above 100 °C is higher than 1 atmosphere, that assumption would imply that the air pressure is negative, a physical impossibility. The dropping of that assumption, however, does not imply any modification of the THM formulation.

4.2.4 Remaining Open Issues - Identification of Gaps in Process Understanding

From a *thermo-hydraulic-chemical* point of view the following remaining uncertainties can be formulated.

The mineralogical transformation of bentonite minerals that are likely to occur at temperatures of relevance (between 100 °C and 150 °C) seem to be limited even over very long times. The fact that in repository relevant conditions, temperatures above a 100 °C coincide with low water saturation might further reduce the already slow reaction rates.

In very low permeability host rocks the time required until full saturation can be considerable. For temperatures above 100 °C drying of (the inner part of) the EBS can occur and vapour movement takes place. As the host rock is saturated to some extent (although not under high hydraulic pressures), geochemical reactions will take place there and be limited in unsaturated regions. The impact of chemistry induced by reactive gases (CO₂) generated in the host rock and moving into the partially saturated bentonite (at elevated temperatures) has not been constrained so far. This includes for example, the impact of suction on thermodynamic equilibria as well as the role of microbiology as partly saturated bentonite might not have developed sufficient swelling pressure yet to suppress bacteria to be active.

From a *thermo-hydro-mechanical* point of view the following remaining uncertainties can be formulated.

Results from several long-term heater-buffer (FEBEX (ENRESA 2006), LOT (Karnland et al. 2009), ABM (Svensson et al. 2011) experiments show minor effects on hydro-mechanical properties after elevated temperature and dismantling such as a decrease in strain at shear failure, a slight decrease in hydraulic conductivity and a slight decrease in swelling pressure. The reason for small deviations observed on for example the hydraulic conductivity after short-term laboratory heating is not fully clear. The impact of the duration of the elevated temperature period on the property change is also not clear. The impact of the duration of the elevated temperature period on the property change is also not clear but changes can occur already after short heating/cooling cycles.

Although the existing data do not point to very significant impacts, one can conclude that a comprehensive view on the impact of temperatures up to 150 °C on THM properties is not yet in place. The data support describing the THM behaviour and supporting the modelling between 100 °C and 150 °C is sparser compared to the support at temperatures lower than 100 °C.

In relation to the above, some impact of vapour on bentonite properties under repository relevant conditions (120 - 150 °C) can be expected and should be clarified taking into account

repository relevant vapour regimes. This impact might be due to minor surface cementation effects although this needs further confirmation.

The extensive modelling performed at the URL scale as part of PEBS highlighted the sensitivity of the modelled temperatures and relative humidities to the evolution of the thermal conductivity especially when the bentonite is at very low saturation. The strong correlation between this relationship and the parameters determining the relative permeability curve for the materials further complicates the analysis and although a reasonably good fit can be achieved, some uncertainty remains. In the column experiments, this sensitivity was less obvious.

It is suspected that vapour movement is scale dependent and plays a more important role in the HE-E experiment (at the URL scale), although also in the column experiments (at the laboratory scale) water distribution in the vapour phase has been observed. At the URL scale vapour movement might enhance the homogenisation of the water contents of the different materials (blocks, pellets) in the HE-E. This might indicate that certain material characteristics evolve during the early hot and dry phase.

Initial dry density differences of the bentonite materials are expected to diminish (e.g. due to the almost unconstrained initial swelling of the blocks surrounded by relatively dry granular material). On the other hand new water content and dry density gradients will develop due to the radial incoming water flux from the tunnel and the outgoing heat flux from the heaters in the centre. A similar partial homogenisation process has also been observed as part of the dismantling of the EB experiment (as described in Case 3; Palacios et al. 2014). In contrast, the development of water content and density gradients due to hydraulic and thermal regime has been observed during the partial dismantling of the FEBEX in-situ test at the Grimsel Test Site in 2002 (Huertas et al. 2006). Part of the developed dry density gradients might disappear when the temperatures decrease. However, it cannot be excluded that some of the heterogeneity will remain locked-in in the long term.

4.3 Case 3: HM Evolution of the Buffer (Focus on Mechanical Behaviour)

4.3.1 Introduction to the Overall Process Understanding

Sealing ability is essential for the engineered clay barriers in all repository concepts. This is normally achieved by development of a swelling pressure and a low hydraulic conductivity. The swelling pressure may also impact the impact the other barriers (canister and host rock) in the repository. The mechanical properties of the installed EBS, which may consist of a mixture of blocks, pellets and engineering voids, will be entirely different from the situation after full saturation. It is therefore important to understand:

1. The mechanical evolution during the saturation phase
2. The final situation after equilibrium

A good knowledge of the mechanical evolution is necessary to ensure that a given design is sufficient to meet the performance targets.

4.3.2 Contributing Issues from Experiments and Experiment Modelling in PEBS

EB Experiment

The EB experiment has already been summarized in Case 1 (see Section 4.1.2.3). Related to this Case 3, the most relevant in situ mechanical information during the hydration process refers to the evolution of the swelling strain and pressure in the barrier. As indicated in Case 1, and according to the monitoring of the experiment during its 10-year life, the following data were obtained:

- In the first 1.5-year period of the experiment, saturation was fairly quick, and total pressures in the barrier as high as 1.7 MPa were recorded (while a maximum swelling pressure of only 1.3 MPa was expected).
- Following this relatively short first-period (1.5 years), along the remaining life of the experiment (~9 years), water intake was very slow (and no additional water was artificially injected after 5 years); and the recorded swelling pressures increased also very slowly, up to a recorded maximum value of 2.2 MPa.

After dismantling the experiment, the visual observations and the on-site and laboratory tests results have confirmed that the bentonite barrier was highly saturated, sealing all the initial voids. Also, the hydraulic conductivity was as low as foreseen, and the thermal conductivity in the expected range. However, even though some degree of homogenization of the bentonite materials has already taken place (the blocks have decreased in density and the GBM has increased in density), there is still a trend for the moisture content to increase towards the bottom of the section (while the dry densities are lower than in the upper part), as it is shown in Fig. 4.7. It is worth noting when analysing these results that, in this specific experiment, a noticeable initial segregation took place due to the GBM emplacement procedure used.

Water content (%), Section B2

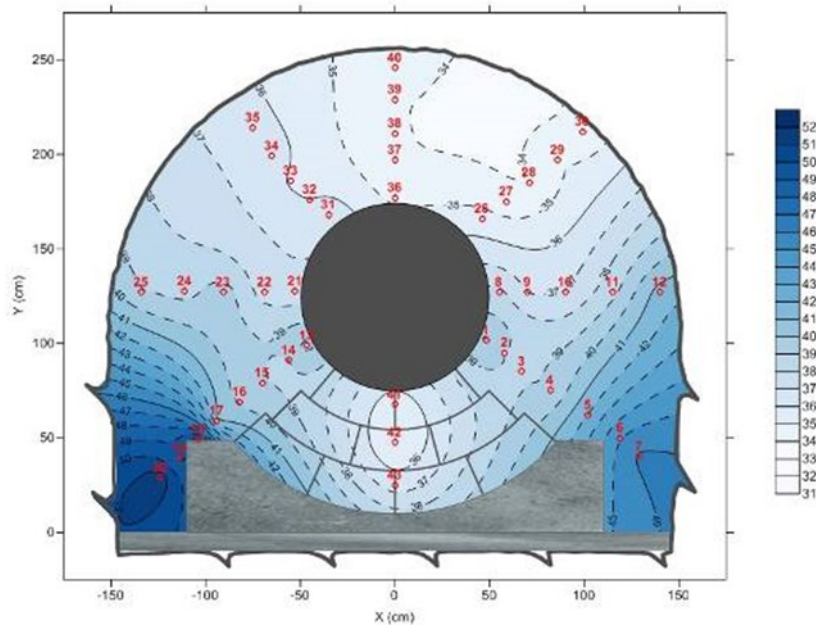
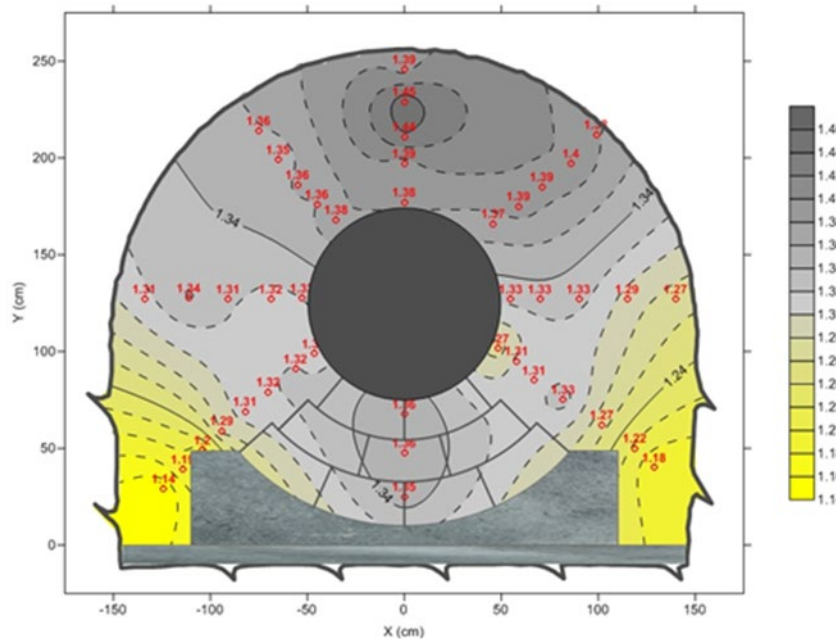
Dry density (g/cm³), Section B2

Fig. 4.7: Water content and dry density spatial distributions in the EB experiment

Contributing Issues from Experiment Modelling

The EB experiment has been modelled using the computer code CODE_BRIGHT. Because the test is run under isothermal conditions, only the hydro mechanical component of the full THM formulation is considered in the analysis. Therefore the balance equations that are solved are i) mass balance of water, ii) mass balance of solid phase, and iii) momentum balance

(equilibrium). Although the material is unsaturated initially, the air mass balance equation is not solved but the gas pressure is considered constant and equal to atmospheric. Note that the solid phase balance equation can be eliminated and the resulting unknowns are water pressures or suctions (hydraulic unknown) and displacements (mechanical unknown). The main hydraulic constitutive equation is Darcy's law that relates water potential gradients to liquid flow taking into account unsaturation via a relative permeability parameter.

An important feature of the experiment is that most of the barrier material is made up of pellets. Therefore, it is a double porosity material where the micro porosity is enclosed inside the pellets and the macro porosity corresponds mainly to the voids between pellets. Naturally, the relationship between micro- and macro porosity changes as the hydration of the barrier proceeds; the pellets swell, micro porosity increases whereas macro porosity reduces, their space being progressively occupied by the expanding pellets. To take into account those phenomena a double structure (porosity) constitutive model has been adopted to describe the mechanical behaviour of the barrier material. The two structural levels (macro and micro) are explicitly considered in the model; the microstructure is governed by a nonlinear elastic law whereas the macrostructure is governed by an elasto-plastic law that encompasses both unsaturated and saturated states. Interaction functions are defined that account for the interplay between the two structure levels. Importantly, permeability for liquid flow is related to the macro porosity and not to the total porosity. Although more information is required to fully define this double structure model, it is believed that it represents better the actual hydro mechanical behaviour of the material. Barrier blocks located below the canister are modelled by a conventional elasto-plastic single structure constitutive model.

The formulation does not include a provision for the possibility that the water in the microstructure might reach density values larger than 1 g/cm^3 . At some point in the experiment, it was thought that this phenomenon could explain the excess of water that entered the test area, but dismantling observations have shown that this phenomenon has not occurred in this experiment.

The numerical modelling of the experiment has included the full period of hydration as well as the dismantling operation. The analysis has tried to simulate, as closely as possible, the hydration conditions that have been applied during the tests. This is not straightforward as there were some episodes in the experiment in which the process of hydration was not fully controlled. Also, there was no information on the amount of water to be attributed to each water entry point; a uniform distribution of water entry among the various hydration points has been assumed.

An example of the prediction of the numerical model for a particular analysis is shown in Fig. 4.8. It can be observed that the results obtained from the modelling are broadly consistent with the dismantling observations. For instance, an important degree of dry density homogenization is obtained between the blocks under the canister and the pellets above the canister. The observed lateral higher porosity of the barrier is also an outcome of the analysis. It is also predicted that the barrier is almost fully saturated throughout, again consistent with observations.

It should be noted that uniform initial conditions of the barrier have been assumed, the observed material segregation during emplacement has not been considered. Therefore, the computed degree of heterogeneity at the end of the test arises solely from the test geometry, initial material distribution and pattern of hydration. Initial non-uniformity of the material would of course lead to somewhat different results but no information on this initial distribution is available. It can thus be stated that the progression of homogenization and the degree of heterogeneity at the end

of the test is adequately represented by numerical modelling. Numerical analysis is therefore a useful tool to predict the resulting heterogeneity in the barriers after hydration, at least over time periods similar to those of the EB test.

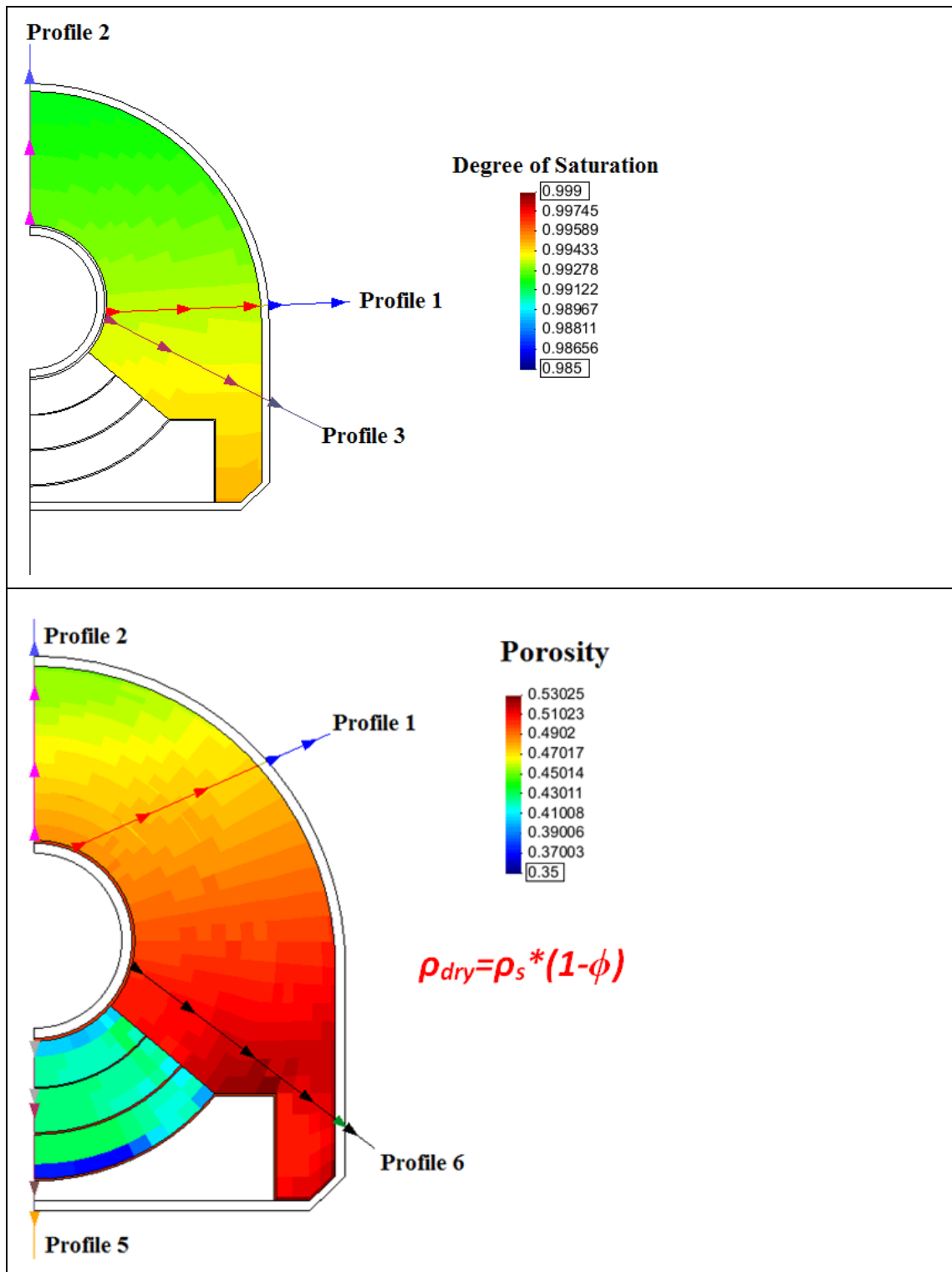


Fig. 4.8: Spatial distributions of computed degree of saturation (top) and porosity (bottom) in the EB experiment

Alternative Approach for Mechanical Modelling

It has been observed in full scale experiments that buffer systems do not become totally homogenized at full water saturation. Causes of the remaining heterogeneity in a buffer system designed according to the Swedish KBS-3V concept, consisting of Wyoming bentonite (MX-80) in blocks (b) and a pellet-filled slot (p), were here studied using an analytical model, developed from a thermodynamic basis. The model solutions were compared with data from a full scale field experiment, the Canister Retrieval Test (CRT), from which information of the initial and final state was available.

The analytical model enabled investigations of how the homogenization at canister mid-height was affected by different wetting scenarios (fast or slow) and different pressure ratios,

$$\alpha = p^p / p^b ,$$

between the pellet slot pressure, p^p , and block pressure, p^b . The analytical model is expressed in terms of a "water retention balance" between the buffer materials,

$$s_{free}^p \left(\frac{\rho_w}{\rho_s} e^p \right) - \alpha s_{free}^b \left(\frac{\rho_w}{\rho_s} e^b \right) = 0 ,$$

where void ratios, e^p and e^b , and density of water, ρ_w , and solid, ρ_s , are present. The water retention curves are given in the form,

$$s_{free}^m (w^m) = \exp(a^m - b^m w^m), \quad m = p, b ,$$

as functions of the water contents, w^b and w^p , and with parameters a^b , b^b , a^p , and b^p , dependent on referential water content. The referential water content is defined as the water content from which absorption or desorption started when the buffer system became fully water saturated, i.e. it will account for hysteresis in the water retention properties.

The analytical model showed that the wetting process (serial = fast or parallel = slow in Fig. 4.9), expressed by use of the known wetting/drying retention curves of the clay, had a significant impact on the level of remaining heterogeneity in the buffer system. Also, the pressure ratio, α , was found to have a significant effect on the homogenization process. Fig. 4.9 shows a compilation of the findings of the analytical study.

The thin solid and hatched lines in the left graph in Fig. 4.9 show combinations of final pellet slot void ratio and final block void ratio obtained from using the analytical model for some given combinations of {wetting process, pressure ratio}. The CRT final state at canister mid-height, indicated by the red circle, is given as a reference point, and the thick black line including this point is obtained from assuming constant volume, i.e. only allowing for radial deformation, which can be considered representative at canister mid-height. When evaluating the effects it therefore seems natural to stay on the constant volume assumption line to restrict the solutions.

It can be seen that when assuming (wetting process = parallel, pressure ratio = 1), indicated by the black hatched line, the pellet void ratio = block void ratio, i.e. perfect homogenization occurs. If still considering parallel wetting but decreasing the pressure ratio, the magnitude of homogenization decreases. The states obtained for $\alpha = 0.9$ and $\alpha = 0.7$, which have been

labelled D and C respectively, are visualized as average final void ratio profiles to the right in Fig. 4.9. As can be seen, the effect from changing pressure ratio is significant.

If the wetting process now changes (i.e. wetting process = serial, pressure ratio = 1) this actually coincides with the CRT state. The choice of pressure ratio, $\alpha = 1$, however, is not considered representative in CRT; the correspondence is rather a coincidence. The states obtained for $\alpha = 0.9$ and $\alpha = 0.7$, which have been labelled B and A respectively, are visualized as average final void ratio profiles to the right in Fig 4.9. If comparing states A and C or states B and D, the effect from changing wetting process from serial to parallel is obtained. As can be seen, the effect from changing wetting process is significant.

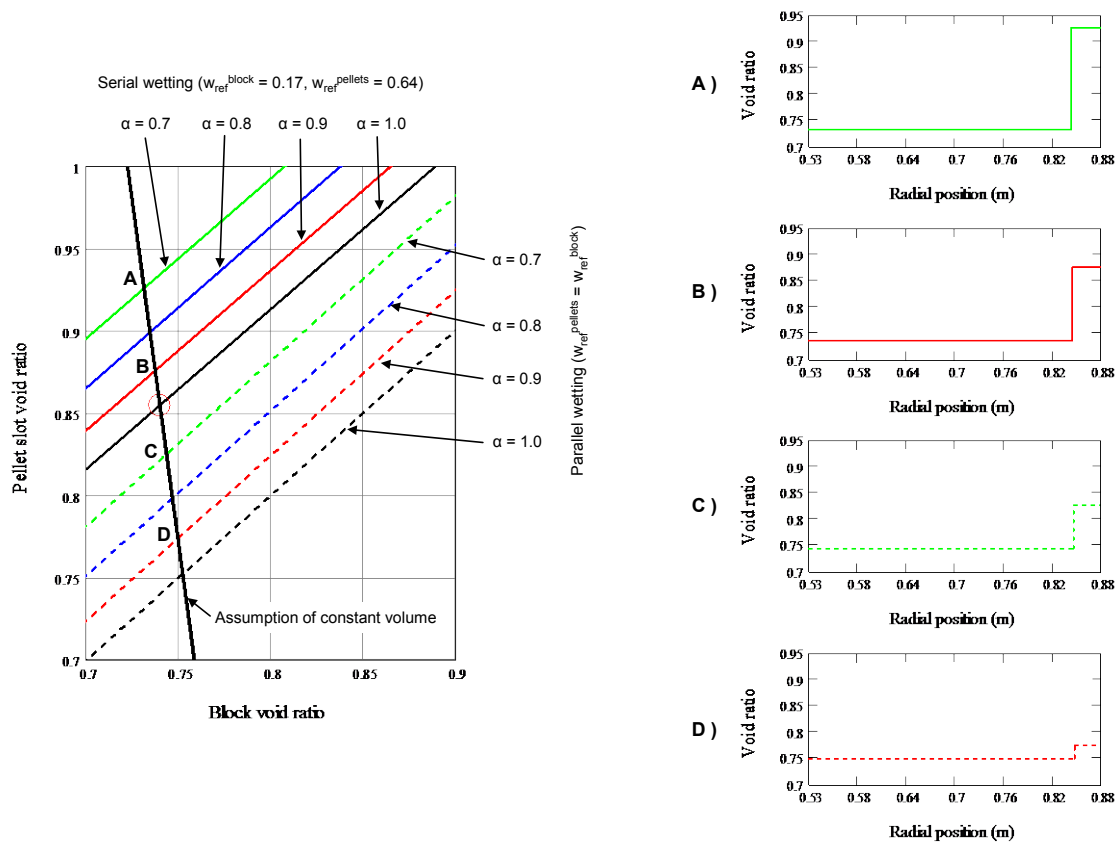


Fig. 4.9: Compilation of the analytical model results for serial = fast (solid lines) and parallel = slow wetting processes (hatched lines) assuming different pressure ratios α

To the right, obtained average void ratio profiles are shown for four different choices, A, B, C, and D, in combinations of wetting process and pressure ratio along the constant volume assumption with the CRT state, indicated by the red circle, as a reference.

The next question is if it is possible to somehow evaluate the analytical model against the CRT data? One way to do this is to feed the model with CRT data and see how well the model describes the wetting process in the experiment when comparing with reality. So, the final void ratios according to the CRT data ($e^b = 0.74, e^p = 0.86$), and a suitable range for the pressure

ratio, $\alpha = \{0.8, 0.9\}$, can be used to produce allowable combinations (in the model's perspective) in referential water contents for the block and pellet slot.

The obtained result shows that the highly idealized parallel and serial wetting are out-of-bounds, and that the allowed wetting process is considerably closer to serial = fast than to parallel = slow wetting. In the CRT experiment the buffer was indeed saturated fast at canister mid-height. After filling the outer slot with pellets at installation, the available pore space between the pellets was water filled using tubes inserted in the slot until a water mirror appeared at top, and during the operational phase, water was fed through filter mats installed at the deposition hole wall. This gave a rapid wetting from the pellet slot side. The model must be said to produce a result, a wetting process close to serial = fast from the pellet slot side, that corresponds well with reality.

The analytical model described above is specially designed for a buffer system consisting of MX-80 block and MX-80 pellet slot where the pellet slot wetting precedes or is concurrent with the block wetting. One could of course alter these premises and derive models on the same thermodynamic basis designed for other geometries, materials, and wetting scenarios.

One interesting case could for instance be to evaluate the homogenization process in the EB in-situ experiment. In Fig. 4.10 an iso-map of dry density obtained from plotting excavation data from the EB in-situ experiment is shown. As can be seen, the density is higher at the top of the tunnel as compared to the rest of the tunnel volume. This is interesting given that, at installation, the buffer blocks indicated below the canister had higher density as compared to the rest of the tunnel backfill composed of granular bentonite. Thus, the buffer system can be thought of as "over homogenized". This experiment used Febex bentonite and the artificially controlled wetting is believed to be initialized in the bentonite blocks below the canister.

If the analytical method is to be utilized for this case, a new model has to be derived specially designed for a new geometry, a new material, and a new wetting scenario (the wetting of the high density material here precedes the wetting of the lower density material).

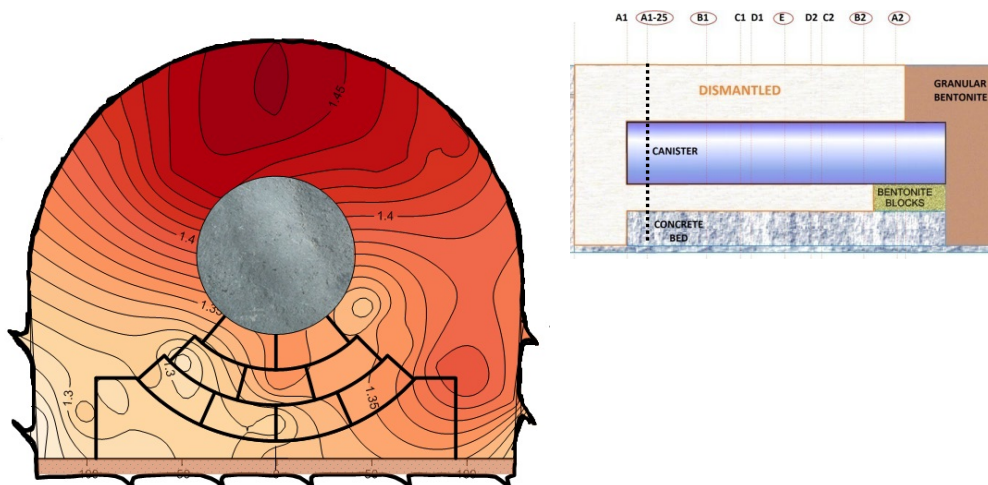


Fig. 4.10: Left, an iso-map of dry density in the EB in-situ experiment obtained at dismantling of the section indicated by the black hatched line to the right

The pictures are taken from Aitemin's presentation held at PEBS annual project meeting in 2012.

FEBEX Mock-up Test

The indicators of a fully-saturated buffer are high stable average values for swelling pressure and relative humidity, a positive trend in fluid pressure values, and very low water intake. All of them are occurring within most of the volume of the barrier, including homogeneous thermal conductivity, but not in the closest zone around the heaters. Although the buffer is practically saturated, full saturation close to the heaters has not been reached yet.

As noted in Section 4.1.2.2, Martin & Barcala (2014), according to Villar and Gómez-Espina (2009), indicate that overall external suction values (around 14 MPa, ~85 % Sr) have remained almost steady during last ten years, some differences can be noted.

In the hydration front, the full saturation for the expected dry density is confirmed by suctions lower than 4 MPa (~23 % wc, ~100 % Sr). This band is more than 0.15 m-thick.

In the heater zone, the presence of non-saturated material is also confirmed by suctions higher than 20 MPa (~18 % wc, ~77 % Sr) and even higher than 41 MPa (~16 % wc, ~68 % Sr) at the middle heater. This "dry" band is 0.05 m-thick but is still going on to saturate.

The saturation greatly affects the thermal conduction through changes in thermal conductivity (related to water intake and/or microstructural changes in the bentonite), which duplicates from the dry state to the fully saturated one (Villar 2002), from 0.6 to 1.3 W/m.K, respectively, with a sharp increase between 40 % and 90 % Sr. After the initial drying effect on the thermal conductivity, most of the barrier presents current thermal conductivity values higher than 1.2 W/m.K, with exception of the closest central zone of the heaters where thermal conductivity ranges between 0.8-1.0 W/m.K.

The higher overall thermal conductivity (averaged increase of 25 %) explains the decrease in the temperatures within the barrier, due to the increased thermal load, and the enhanced influence of the seasonal temperature variations inside the barrier. The influence of thermal transport is shown in the observed fluctuations in relative humidity, total pressure and fluid pressure values.

Two different processes could explain the strong coupling observed between the temperature in the barrier and the hydro-mechanical (HM) processes, depending on the local saturation of the barrier: the thermal expansion of the phases in the outer full-saturated zone and the suction changes, probably in combination with the previous one, in the inner less-saturated zone.

The large-scale long-term major experimental observations, related to HM processes, are the same as those described in the case 1:

- Experimental measures confirm transient effects of evaporation (drying) and condensation (wetting) and the migration of water vapour through the barrier, even at medium-high degrees of saturation. Both, water inflow and water vapour outflow are mainly radial and homogeneous.
- The transport processes are controlled by the hydraulic properties of the bentonite and affected by absolute temperatures and/or thermal gradients imposed by heating (directly related to the disposal system through the barrier geometry and boundary conditions; Villar et al. 2012).
- The pressure values become homogeneous and stable as the buffer material saturates, reacts and changes its microstructure. Values for the different directions are converging to the swelling pressure values measured in the laboratory, for the expected dry density of the bentonite.

From above, the buffer achieves the required performance over time as saturation increases, as shown by the sufficient and homogeneous swelling pressures developed in the highly saturated outer layer of the buffer and the homogeneous high thermal conductivity achieved. Besides, FEBEX mock-up test supports the evidence of the slowing down of hydration at the later stages, under a specific thermal load and geometry, and the implications of the thermal aspects on the transport processes.

4.3.3 Key Issues Identified during PEBS Representing Uncertainties related to Case 3

In summary some of the findings from PEBS Case 3 are:

- The homogenization of a bentonite buffer is efficient; even it is installed as a mixture of high density blocks and low density pellets. The EB experiment has confirmed that a highly saturated bentonite barrier was able to seal all the initial voids.
- A swelling pressure can develop even in a partially saturated buffer. In Febex Mock up, the buffer achieves the required performance over time as saturation increases, as shown by the sufficient and homogeneous swelling pressures developed in the highly saturated outer layer of the buffer.
- Numerical analysis is a useful tool to predict the resulting heterogeneity in the barriers after hydration. For the EB experiment; it can be observed that the results obtained from the modelling are consistent with the dismantling observations.
- Models have indicated that the final heterogeneity of mass in a bentonite barrier may depend on the wetting history.
- It is still unclear how much of the heterogeneity will gradually disappear. Nonetheless, even with the observed variations in density the barrier still provides effective self-sealing and sufficiently low hydraulic conductivity.
- Heating will affect the mechanical properties of bentonite, however the effects are small below 100 °C.

In summary, the results obtained in PEBS confirm the assumed mechanical performance of a bentonite barrier in geological disposal. The uncertainties in the long-term performance of bentonite barriers have thus been reduced in some areas.

4.3.4 Remaining Open Issues - Identification of Gaps in Process Understanding

There will be remaining heterogeneities in a bentonite buffer after full saturation. The main reason for this is the heterogeneities in the installed material, i.e. blocks, voids and pellets, but the wetting history may also have a significant impact. It is still unclear how much of the heterogeneity, if any, in bentonite density that arise due to differential wetting rates will gradually disappear over time.

This will have an impact on future assessments, since it may be difficult to define a single swelling pressure for a heterogeneous buffer. Thus, the effect of the heterogeneities has to be considered in the evaluation of the fulfilment of the safety functions.

4.4 Case 4: Geochemical Evolution

4.4.1 Introduction

The focus of Case 4 was on the geochemical evolution at the interfaces between a bentonite buffer and adjacent materials, namely the steel canister¹ and, in case of a repository in argillaceous rock, the concrete lining. At these interfaces chemical reactions leading to alterations of the EBS and also the canister/concrete are unavoidable. These may depend on temperature, pore water composition (granitic water or argillaceous type water), and alteration progress. The most relevant questions with respect to EBS performance in this context are:

- What is the degree of alteration, what are the properties and thicknesses of the altered zones?
- How does the alteration affect the overall buffer performance, in terms of swelling pressure and hydraulic behaviour?

Several experiments representing different concepts and stages of repository evolution were performed within PEBS. The set-up and experimental conditions are summarized in Section 4.4.2. The experimental outcomes and the related numerical modelling performed for investigating the capability of thermo-hydraulic-chemical models including porosity change (THCm) are presented in Section 4.4.3, together with results obtained outside PEBS for comparison. The findings were used for predicting the long-term evolution of repositories in granite or clay following the Spanish concepts as presented, again in the context of results of comparable previous work outside PEBS, in Section 4.4.4.

The information obtained was categorized into a set of key issues regarding the geochemical evolution at the interfaces. These comprise the potential bentonite alteration at temperatures above 100 °C which is also discussed in Section 4.2, uncertainty in the bentonite hydration rate stemming from pore clogging, uncertainties in the chemical reactions and parameters for modelling, and the issue of the time-scale of some geochemical reactions, all of which are discussed in detail in Section 4.4.5. Finally, the uncertainties are evaluated and remaining issues are summarized in Section 4.4.6.

4.4.2 Contributing Experiments in PEBS

Geochemical experimentation concentrated on investigation of the processes at the interfaces canister/bentonite (for a repository in granitic rock) and bentonite/concrete (for a repository in argillaceous rock). Two stages in repository evolution were distinguished:

5. Later post-closure phase, full saturation of the EBS, anoxic conditions: Respective experiments were performed in small cells.
6. Operational, early post-closure and transient phases of the repository, partial saturation and oxic to suboxic conditions: Respective experiments involved medium cells.

¹ The case of alteration as result of corrosion of a copper canister is not treated, as the effects are negligible.

4.4.2.1 Small Cell Experiments

The so-called small cell experiments were designed to reproduce the repository conditions prevailing 1,000 to 3,000 years after emplacement of the waste, when the bentonite is fully saturated.

For a repository in granite (NF-PRO project; details in Torres et al. 2007, 2008); the interface FEBEX bentonite/iron powder (or C-steel plates) was studied and the system was hydrated with granite type water collected at the Grimsel Test Site (GTS).

For a repository in an argillaceous formation (PEBS project; details in PEBS D2.3-3-2, Cuevas et al. 2013); the interfaces mortar/FEBEX bentonite/magnetite were studied and the system was hydrated with a synthetic argillaceous type water. A lime mortar-compacted bentonite interface was placed at the hydration zone. The lime mortar was used to provide a source for calcium and alkalinity. A magnetite-compacted bentonite interface was placed on the opposite side to hydration. Magnetite was used as the prevailing corrosion product at the time scale considered. Natural FEBEX bentonite and FEBEX bentonite depleted in magnesium (artificially prepared by modifying the distribution of exchangeable cations, based on modelling, Samper et al. 2010) were used. The experiments ran isothermally at 60 °C (based on modelling, ENRESA 2004). Hydration was made with reference synthetic clay water (pH 7.5, Na⁺-Ca²⁺-SO₄²⁻ type water). The thickness of the bentonite was selected so that the corrosion and hyper alkaline fronts would interact within the time span of the experiment (18 months).

4.4.2.2 Medium Cell Experiments

Experiments in the medium cells were performed to simulate the operational, early post-closure and transient phases of the repository, i.e. the state before achieving full saturation of the EBS, characterised by oxic to suboxic conditions. The experiments considered the interface between FEBEX bentonite and iron powder with hydration by granite type water collected at the Grimsel Test Site (for a repository in granite) and the interface between concrete and FEBEX bentonite with hydration by synthetic argillaceous type water (for a repository in an argillaceous formation). The experiments were designed to establish the evolution, through sequential dismantling at different times from 6 months to 7 years, of the chemical processes from the unsaturated to the saturated conditions, both at the concrete/compacted bentonite and the iron/compacted bentonite interfaces separately. Therefore, a number of identical experiments were assembled simultaneously already by mid-2006 in the frame of the NF-PRO project and were sequentially dismantled (details in PEBS D2.3.3-1, Turrero et al. 2011).

Hydration was realised through the concrete in the cells with the concrete/bentonite interface. The hydration solution was reference synthetic clay water (pH 7.5, Na⁺-Ca²⁺-SO₄²⁻ type water). The cells with the iron/bentonite interface were hydrated through the bentonite. The hydration solution was granitic water collected at the Grimsel Test Site (pH 9.7, Na⁺-Ca²⁺-HCO₃⁻ type). The heat source (100 °C constant temperature) was located on the opposite side to hydration. A constant gradient of temperature between the hydration and the heat source of the cells was maintained by means of a container in which water circulated at room temperature (25 °C). The concrete was sulphate-resistant ordinary Portland cement (CEM I-SR) following the recipe by CSIC-IETcc CEM-SR1. Iron powder with a particle size of 60 µm and purity higher than 99 % was used for the experiments. Iron powder was used to enhance the formation of corrosion products at the interface.

4.4.3 Experimental Data and Interface Modelling

4.4.3.1 Iron Interfaces

Experimental data from CIEMAT's corrosion tests indicate that there is a sequence of corrosion products, under saturated and reducing conditions. Ferrous hydroxide is the first reaction product, then magnetite is the most stable product, reached via mixed Fe(II) and Fe(III) iron hydroxides, including green rust-Cl. Iron hydroxides precipitate near the interface. No newly-formed iron-rich clay phases and no mineralogical alteration of the bentonite were detected in experiments lasting two years. The corrosion rates under these conditions estimated from C-steel plates were approximately 2 $\mu\text{m/a}$.

Under the transition from unsaturated aerobic to anaerobic conditions as reproduced in medium cell experiments the reaction pathway for iron corrosion starts with the formation of iron hydroxide ($\text{Fe}(\text{OH})_3$), then lepidocrocite and goethite (oxy-hydroxides), and finally hematite and maghemite (Fe_2O_3) when the system is depleted in oxygen.

At the present state of knowledge, the extent of bentonite alteration due to iron corrosion, and the exact type of alteration products produced, cannot be predicted with any confidence. The conversion to a non-swelling 7A mineral might be slow (Bradbury, in prep.). Other effects have to be considered as well, reduction of the porosity at the interface due to precipitation of Fe-corrosion products, competition for sorption sites, reduction of the Fe^{3+} in octahedral position (Bradbury, in prep.).

Prediction of the potential behaviour of smectite in the presence of a steel container and their related alteration products, under nuclear waste repository conditions, is made difficult by the likely temperature range of 25 - 90 °C. Under such low temperatures, very little mineralogical change could be determined from experiments carried out over short durations (<2 years) and for liquid/mineral ratios typical of an engineered barrier (<0.1years). According to Mosser-Ruck et al. (2010), low temperatures (<150 °C) and large liquid to solid and Fe/bentonite ratios seem to favour the crystallization of the serpentine group minerals instead of Fe-rich trioctahedral smectites or chlorites, the latter being favoured by higher temperatures. The role of Fe/clay (Fe/C) and liquid/clay (L/C) ratios and the competition between them at different temperatures is a crucial point in understanding the transformation of smectite in contact with metallic iron. Experiments carried out at higher temperatures could, however, be extrapolated to lower temperatures as the type of mineralogical changes observed at high temperature is predicted by numerical modelling either at high (300 °C) or much lower temperatures (80-100 °C). However, kinetic predictions using constraints from temperature-time pairs still require experimental calibration.

The iron alteration zone under unsaturated aerobic to anaerobic conditions in experiments lasting 7 years has an average thickness of 1 micron and the Fe-influenced zone was estimated to be 20 μm . The alteration zone consisted of a mixture of corrosion products (nanoparticles) and bentonite enriched in chloride and sulphate, probably due to the compression effect of bentonite swelling on Fe powder. No newly-formed Fe-rich clay phases were identified.

Under saturated and iron-reducing conditions a redox front was generated in the first 7 mm of the bentonite block and a thick layer of ferrous hydroxide precipitates on the surface of the bentonite (Torres et al. 2007, Torres et al. in prep.). Non-structural iron was around 5 %wt. just at the interface (1 mm), filling the voids in compacted bentonite, 0.1 %wt. and 0 %wt. at 3 mm and 5 mm from the interface, respectively. No iron was found in the exchange complex of the bentonite (<0.5 meq/100 g). Regarding these conditions, BET SSA measured at the interface

decreased by 20 %, at 3 mm from the interface by 5 % and after 5 mm, the SSA found was in the range of the ones measured for natural FEBEX bentonite. Thus, a minimum impact on physical-chemical properties of the bentonite is expected beyond a few mm distance.

The corrosion tests carried out by CIEMAT have been modelled by UDC with coupled THCM models. Model results of the small experiments indicate that (Samper et al. 2013a, 2014a, Deliverable 3.5-4):

1. Magnetite and $\text{Fe}(\text{OH})_2(\text{s})$ precipitate and compete for Fe^{2+} precipitation. These corrosion products penetrate a few mm into the bentonite.
2. Fe^{2+} is sorbed by surface complexation.
3. Fe^{2+} cation exchange is less relevant than Fe^{2+} sorption.
4. The numerical results fit the measured data of the iron weight for the tests performed at 25 °C, 50 °C and 100 °C.

4.4.3.2 Concrete Interfaces

The diffusive transport of solutes across a cement-clay interface leads to sharp gradients in pH (and pCO_2). Such sharp gradients encourage rapid precipitation of carbonates, hydroxides and CSH phases, leading to decreased porosity (e.g. Watson et al. 2009, PEBS D2.3.3-2). Fast protonation-deprotonation reactions at clay edge sites can typically neutralise 55 moles of hydroxyl ions per cubic metre of MX-80 bentonite (at 2000 kg m^{-3} water-saturated compaction density) (Savage et al. 2007). In addition, montmorillonite and other minerals present, such as quartz, feldspars, pyrite, and gypsum, are slowly dissolved at high pH. Such reactions consume hydroxyl ions and slow down the progress of the high-pH front.

Besides the high-pH front, cation exchange reactions are taking place. These cation exchange reactions advance in front of dissolution-precipitation reactions (e.g. Fernández et al. 2009a and b). As a consequence of this process the exchange of Na^+ in MX-80 bentonite by K^+ and Ca^{2+} from cement leads to a decrease in montmorillonite swelling pressure at the concrete interface. Secondary minerals such as clays, hydroxides, carbonates, calcium silicate hydrates, and aluminosilicates such as zeolites and feldspars (e.g. Savage et al. 2007, Fernández et al. 2010, Savage et al. 2013) are formed in a zonal fashion. C(A)SH, illite, feldspars, and the more aluminous zeolites are preferentially formed at very high pH (>13) (Fernández et al. 2013), i.e. in regions close to the interface, and the more siliceous zeolites form at lower pH, i.e. in regions further away from the interface. However, the formation of zeolites and presumably other secondary phases is not easily observed when bentonite dry densities up to 1.6 g/cm^3 are used in the experiments (Fernández et al. 2013).

The study of concrete-bentonite interfaces in a real OPC high-pH (medium cells) concrete or lime mortar (small cells) using a saline clay formation porewater in the hydration source produced complex reaction paths and small evidence of the above described high-pH typical minerals. When the concrete interface was hydrated (up to 6.5 years in the medium cell experiments of PEBS) with a saline Mg, Na-sulphate-rich water containing chloride and bicarbonates (simulating a reference clay formation porewater), carbonates and brucite precipitated. Concrete acts as Mg and inorganic carbon filter. Inside the concrete the reaction path follows the dissolution of portlandite (absent in the cement matrix at long term) and precipitation of CSH (of low C/S (0.8) ratio), calcite and ettringite (near the bentonite interface). Calcite precipitation is more important towards the bentonite, indicating that the carbon source is coming from the bentonite. Finally the concrete/bentonite interface shows the precipitation of

gypsum, CASH gels, MSH gels and carbonates (calcite and aragonite), affecting <2 mm thickness of bentonite. No newly formed zeolites were observed.

Both Dauzères et al. (2014) and Jenni et al. (2014) have shown recently, for low-pH and high-pH concrete interfaces, the significance of carbonation and sulphate phase dynamics in producing porosity changes. Dissolution-precipitation and the associated clogging fronts of calcite (both in concrete and clay) or neo-precipitation of ettringite (concrete) in <1 mm depth will favour local equilibrium as far as the porosity will be reduced locally, which stops or slows down further mineralogical alteration. These cementing processes are more active in high pH OPC than in low pH OPC. However, the slow kinetics of mineralogical alteration might prevent the complete reaction of OPC concrete with bentonite (Savage et al. 2010a). Of course, more information on this aspect will be available when in situ long-term (i.e. 20 years) experiments with real interfaces can be studied.

Alteration thicknesses measured in concrete-bentonite experiments designed in PEBS depend on both the previous alteration of bentonite and the duration of the experiment. Carbonation, produced from the calcite dissolution in the bentonite and its reaction with portlandite is driven by the $p\text{CO}_2$ and pH gradients. This reaction prevailed in relatively long-term experiments (medium cells) and affected <2 mm thickness in the bentonite. On the other hand, CSH-like phases characterize short-term and small size experiments. Mg-depleted (typical of alkaline conditions) bentonite seems to present less buffering potential than the natural clay as a consequence of Mg reactivity and MSH precipitation (Cuevas et al. 2014). Typical bentonite thicknesses affected by mineralogical alteration are within 5-1.5 mm. The role of carbonation as a relevant process in the long term has to be considered as an attenuation factor in potential affection to the main bentonite safety functions.

As has been already mentioned, the concrete alteration effect on bentonite can also be followed in terms of producing changes in the redistribution of exchangeable cations. Considering this aspect in the case of FEBEX bentonite the alteration thickness is larger and it is characterized by drastic magnesium depletion due to its precipitation in alkaline pore water. This effect can be followed 20 mm into the bentonite from the concrete interface in the medium scale cells. In the small cell experiments Mg is completely depleted from the overall 20 mm bentonite thickness when lime mortar is present. In the case of MX-80 Na replacement by Ca should be expected.

UDC has modelled the small and medium lab experiments involving bentonite-concrete interfaces (Samper et al. 2013a; 2014a; Deliverable 3.5-4). The coupled THCM numerical models of these tests capture the main observed trends of mineral dissolution-precipitation. The HB4 models, however, do not reproduce the ettringite and CSH precipitation. For these phases, the numerical model predicts much less precipitation than the observed values. In the 2I3 cell model, there are some discrepancies, especially for ettringite in the mortar and in the bentonite (precipitation is observed in the test), and for brucite precipitation at the mortar-bentonite interface and in the bentonite (not observed in the test). The lack of thermodynamic data for CASH phases prevented UDC to account for CASH phases in their models.

4.4.4 Long-term Modelling

4.4.4.1 Long-term Modelling of a Granite Repository

The long-term interactions of corrosion products and bentonite in a repository in granite were evaluated with a THC model by UDC. The precipitation of corrosion products close to the canister could decrease significantly the porosity of the bentonite and even clog the bentonite pores. The thickness of the bentonite zone affected by the decrease of porosity increases with time. A thickness of 7 cm is reached after 1 Ma (PEBS Deliverable 3.5.3; Samper et al. 2014b). Several sources of uncertainty were analysed with the model by means of sensitivity runs. These runs were performed for the following parameters:

1. The corrosion rate
2. The effective diffusion coefficient of the dissolved species in the bentonite
3. The water flow at the bentonite/granite interface
4. The cation exchange selectivities
5. The chemical compositions of the bentonite and granite porewater compositions

The results of the sensitivity runs show that:

1. The larger the corrosion rate, the faster the porosity reduction close to the canister but the smaller the thickness of affected bentonite (PEBS Project Deliverable 3.5.3; Samper et al. 2014b).
2. The larger the diffusion coefficient, the larger the thickness of the bentonite where magnetite, siderite and calcite precipitate. The penetration of corrosion products varies from 4 to 9 cm in the considered range of diffusion coefficients. The larger the diffusion coefficient the larger the zone of bentonite affected by pore clogging (PEBS Project Deliverable 3.5.3; Samper et al. 2014b).
3. The precipitation of the corrosion products is very sensitive to the groundwater flow Q in the granite. An increase in Q leads to a decrease of magnetite precipitation and an increase of siderite precipitation. The larger the Q the smaller the thickness of bentonite in which precipitation of corrosion products takes place (PEBS Project Deliverable 3.5.3; Samper et al. 2014b).

The thermal transient (accounting for the temperature effect on chemical reactions) produces a decrease in pH and silica dissolution/precipitation. Magnetite precipitation is almost similar to the isothermal case. Therefore, the thermal transient does not have a significant effect on the overall geochemical evolution of the EBS and does not affect the thickness of the altered bentonite (PEBS Project Deliverables 3.5.3 and 3.5.4, Samper et al. 2014b).

The corrosion rate may depend on temperature and other environmental chemical conditions. Experimental evidence is not conclusive regarding the increase of corrosion with temperature. According to the experimental data compiled by King (2008) there is no significant temperature dependence of carbon steel corrosion in contact with bentonite. Some authors, however, claim that the dependence of corrosion on temperature can be accounted for with the activation energy (Féron et al. 2008). UDC has performed model simulations which assume that the corrosion rate increases with temperature. Model results show that the precipitation of magnetite increases slightly and the porosity reduction are slightly intensified when the effect of temperature on corrosion is accounted for. Therefore, it can be concluded that the uncertain effect of

temperature on the corrosion rate does not play a major role in the long-term evolution of the system (PEBS Project Deliverable 3.5.4; Samper et al. 2014b).

Accounting for the dependence of the corrosion rate on the chemical conditions (pH, Eh, Fe concentration) leads to an important change in the patterns of the corrosion products. The thickness of the altered zone is doubled (PEBS Project Deliverable 3.5.4; Samper et al. 2014b).

The long-term effect of the kinetic dissolution of smectite on bentonite porosity and on the thickness of altered bentonite is small. A kinetically-controlled smectite dissolution rate from Rozalen et al. (2008) leads to analcime precipitation. Model results indicate that this reaction is not relevant because only 0.2 % of the smectite dissolves after 1 Ma. Smectite dissolution has the beneficial effect of leading to a slight decrease (1 cm) of the thickness of the altered bentonite zone (PEBS Project Deliverable 3.5.4; Samper et al. 2014b).

Lab corrosion tests performed under anoxic conditions indicate that $\text{Fe}(\text{OH})_2(\text{s})$ and ultimately magnetite are the main corrosion products. Lab tests indicate that magnetite precipitation may have a kinetic control. The kinetic rate law derived from lab tests was used to perform long-term simulations. Model predictions are sensitive to the kinetic law of magnetite precipitation and to the reactive surface area used for magnetite. The thickness of the altered zone decreases when kinetics is considered (PEBS Project Deliverable 3.5.4).

Similar to other reactive transport models of canister corrosion, the UDC model assumes that the canister has the same transport properties as the bentonite. The uncertainties caused by this simplifying assumption should be evaluated in the future. The model could be improved by considering different types of waters: internal, external and free waters.

The numerical results of the UDC models of the interactions of iron and bentonite have been compared to the results reported by Bildstein et al. (2006), Wersin et al. (2008), Savage et al. (2010b) and Marty et al. (2010). Savage (2013) describes how iron-bentonite interactions and the effects of corrosion products on the bentonite were first identified by Nagra in the mid 1980's. ANDRA and JAEA performed experiments and modelling studies in the late 1990's. In the middle of the last decade Posiva and SKB followed with reviews and experiments as part of their KBS-3H research programme. Some of these experiments have been interpreted using coupled THC models (Montes-H et al. 2005, Bildstein et al. 2006, Hunter et al. 2007, Wersin et al. 2008, Samper et al. 2008, Savage et al. 2010b, Marty et al. 2010, Lu et al. 2011). Bildstein et al. (2006) modelled the interaction of the canister with the bentonite and the clay formation in the French reference system with a 1D purely diffusive model under full saturation conditions. The model accounts for a 7 cm thick stainless steel canister, an 80 cm thick MX-80 bentonite-based EBS, and a 10 m long geological barrier of clay host rock. The reaction-transport code CRUNCH was used to investigate the iron-clay interactions at 50 °C over 10,000 years. A constant corrosion rate of 4.3 $\mu\text{m}/\text{y}$ was considered. The potential corrosion products included iron oxides and hydroxides, iron carbonates, and iron-rich smectite and serpentine minerals (Fe-phyllosilicates). The porosity was updated according to the net volume balance of dissolved and precipitated minerals. The model considered a gap at the interface between the EBS and the clayey formation. They performed calculations for two systems, i.e., one containing a bentonite-based EBS and another one without EBS, since both options were considered for the disposal of different types of waste packages. Their results for the system involving bentonite show that pH at the bentonite-container interface increases progressively from 7.7 to a 10.5 after 5,000 years. At this time, pore clogging takes place at this location. When porosity vanishes, the pH increases to values around 11. High values of pH were obtained because no iron hydroxide minerals precipitate while the oxidation of Fe by water produces H^+ and OH^- ions. The Ca-montmorillonite and Na-montmorillonite tend to dissolve in the EBS at both the interfaces with

the canister and the argillite, whereas illite remains stable. In this system the corrosion products and interactions with clay were essentially located in the cell representing the canister. The corrosion products include magnetite (35 %) and cronstedtite (40 %, an iron-rich serpentine). The precipitation of chamosite and siderite are less than 1 %. Porosity clogging occurs in the cell containing the iron after 5,000 years in the case with EBS and after 16,000 years in the case without EBS. Clogging is due to the precipitation of magnetite and cronstedtite, and to a lesser extent, of chamosite and siderite. The different timing is due to the fast precipitation of cronstedtite in the EBS. Porosity is also increasing inside the EBS and the argillite. In the case with EBS, the increase of porosity at the interface with the canister (+6 %) affects around 10 cm of bentonite and is due to the dissolution of the initial Na-montmorillonite and Ca-montmorillonite in the EBS which is not compensated by the precipitation of other minerals. In the argillite, a front develops and reaches 4 m depth into the system after 5,000 years, featuring a +5 % porosity increase with the complete dissolution of Ca-montmorillonite and Na-montmorillonite, and the dissolution of 5 % of illite. These minerals are partly replaced by minerals incorporating iron (up to 12 % of cronstedtite, 2 % of chamosite, and 2 % low-Fe smectite, 0.5 % of siderite) and calcium (up to 10 % of scolecite, a Ca-zeolite). Behind this first front, a small amount of Mg-saponite (0.1 %) also precipitates. Overall, the results of UDC calculations share many features of the results of Bildstein et al. (2006). Magnetite is the main corrosion product while siderite precipitation is much smaller than magnetite precipitation. The reduction of bentonite porosity due to mineral precipitation near the canister/bentonite interface could result in the clogging of the bentonite pores. The bentonite thickness affected by porosity reduction increases with time and it is of ~7 cm after 1Ma. Cronstedtite precipitates near the canister/bentonite interface.

Wersin et al. (2008) studied the impact of iron components released from the corrosion process on the bentonite buffer (MX-80) within the KBS-3H concept using the Olkiluoto site in Finland as a test case. At the front end, a constant concentration boundary condition, corresponding to the porewater in equilibrium with the corroded steel supercontainer, is imposed. At the back end closed boundary conditions are assumed. They performed reactive transport simulations using Crunch Flow with a 1D diffusion model considering site-specific geochemical data from Olkiluoto, iron corrosion, cation exchange, protonation/deprotonation and Fe(II) surface complexation and mineral dissolution/precipitation including thermodynamic and kinetic clay minerals. A general conclusion from this modelling study is that the extent of the bentonite zone transformed to non-swelling material is likely to remain spatially limited (a few centimetres) for very long times. UDC calculations agree with the general conclusion of the results of Wersin et al. (2008).

Savage et al. (2010b) reported a model of the iron-bentonite interactions based in natural analogues. They claim that the studies of the natural systems suggest that the sequence of the alteration of the clay by Fe-rich fluids may proceed via an Ostwald step sequence. They used the code QPAC which was modified to incorporate nucleation, growth, precursor cannibalisation, and Ostwald ripening to address the issues of the slow growth of the bentonite alteration products. They modelled iron corrosion (corrosion rate of ~2 $\mu\text{m}/\text{year}$) and the alteration of MX-80 bentonite in a typical EBS environment. Their model neglected protonation-deprotonation reactions at clay edge sites and cation exchange reactions. They considered two model cases; one with fixed reactive surface areas for secondary minerals; and one with time-dependent variation of the surface areas of secondary minerals. Simulations with fixed mineral surface areas show that berthierine dominates the Fe-bearing minerals, with siderite replacing it for $t > 10,000$ years. Further away from the iron canister, berthierine continues to precipitate in the clay as the Fe^{2+} ions penetrate deeper into the bentonite with time. Large amounts of dolomite also appear, due to the relatively high concentrations of calcium in the host rock groundwater and the release of Mg^{2+} ions from the dissolving Ca-montmorillonite.

This, along with the growth of iron-bearing minerals, results in complete pore blocking near the canister and causes the simulation to end prematurely. The pH of the bentonite pore fluids shows a rapid decrease, such that after 10 years pH decreases from ~10.4 to a value between 6 and 7, the precise value depending upon the location within the bentonite. This pH buffering is due to silicate precipitation reactions. Porosity blocking due to mineral precipitation also leads to a slowing of the steel corrosion rate. The simulations with time-dependent mineral surface areas show the following sequence: magnetite-cronstedtite-berthierine-chlorite. The evolution of the secondary minerals is somewhat different to that predicted by the fixed surface area model. Near to the corroding canister, at first magnetite is the dominant Fe-bearing mineral; this is followed by cronstedtite, which dominates between ~0.04 years and 500 years. Berthierine is again present throughout, but in reduced quantities compared with the fixed surface area model; after 500 years cronstedtite dissolves away and berthierine is the dominant Fe-bearing mineral. Eventually, after about 5,000 years, chlorite grows in, using the berthierine surface growth sites rather than nucleating directly. Further into the clay, there is much less Fe-based mineral precipitation than in the fixed surface area case, with only small amounts of berthierine present. The evolution of pH with time is similar to the fixed surface area case, such that pH decreases rapidly over 1 - 10 years from 10.4 to 6-7. The results of the UDC model cannot be compared directly to those of Savage et al. (2010b) because the UDC model considers neither the Ostwald step nor the time-dependent variation of surface areas of secondary minerals.

Marty et al. (2010) performed a model of the long-term alteration of the engineered bentonite barrier in an underground radioactive waste repository. The study was focused on the possible feedback effects of geochemical reactions on the transport properties (porosity and diffusion) of a compacted MX-80 bentonite. The system was modelled in reducing conditions by using the KIRMAT code. The model considers a 1 m thick buffer of MX-80 bentonite which is in contact on one side with a geological fluid (Callovo-Oxfordian groundwater at 25 °C) diffusing into the engineered barrier. The calculations assumed a constant temperature of 100 °C in order to estimate a maximum thermal effect on the mineralogy of the engineered barrier. By considering the temperature and the long-term evolution of the system, the mineralogical transformations were considered as being more important than the surface complexation and cation exchange reactions. As a consequence, these reactions were neglected in the model. After 100,000 years of simulated mass transport-reaction, the model predicted mineralogical modifications of the EBS in contact with the geological interacting fluid and with Fe²⁺ ions provided by the corrosion of the steel overpacks. According to the degree of smectite transformations the mineralogical modifications of the MX-80 bentonite until 100,000 years showed three distinct zones: (1) The first zone results from the mass transport of the geological groundwater through MX-80 bentonite. The resulting alteration front corresponds to a strong illitization of the montmorillonite together with the precipitation of quartz, saponite and vermiculite. (2) In the middle of the EBS, the volume of montmorillonite remains quite constant. The saponitization and the illitization processes can be distinguished in lower proportions as minor phase. (3) The third alteration front is constituted by significant precipitations of Fe(II)Al-chlorite, Fe(II)-saponite and magnetite in contact with the steel overpack. The precipitation of these phases decreases significantly the porosity of the EBS. Marty et al. (2010) concluded that only the outer parts of the simulated system are significantly affected. The dissolution of the montmorillonite contained in the MX-80 bentonite is mainly observed in the zone influenced by the groundwater mass transport and partly in a zone adjacent to the container. Despite the neo-formation of illite, vermiculite, saponite and magnetite detected in the barrier, the predicted evolution of the porosity is limited in the main part of the EBS and the porosity even tends to decrease in the perturbed outer zones. A porosity clogging near the steel overpack significantly decreases molecular diffusion. As a feedback effect, the influence of the iron corrosion on the EBS mineralogy then decreases as a function of time due to the reduction of the bentonite porosity. The long term transformation of the bentonite should have only a minor influence on

the amount of montmorillonite. In the modelling, more than 60 % of the initial montmorillonite is preserved after 100,000 years, and additional swelling minerals such as vermiculite and saponites are formed. Marty et al. (2010) recognize that the calculation may overestimate the transformation rate of the minerals because they assumed a fixed maximum temperature of 100 °C throughout the simulation time. The UDC model is more realistic because it takes into account the time variability of temperature for the geochemical simulation.

As a summary, the conclusions of the simulations performed by UDC within the PEBS project are consistent for the most part with those reported by others for similar systems. In some cases, however, there are marked differences in the geochemical systems and the hypotheses used by the modelling teams. This is especially the case for the simulation of Savage et al. (2010b).

4.4.4.2 Long-term Modelling of Clay Repository

UDC performed long-term THCM simulations for a repository in clay by using an axisymmetric two-dimensional model. Mineral dissolution/precipitation is especially marked in the concrete (PEBS Project Deliverable 3.5.4; Samper et al. 2014c). Kinetically-controlled smectite dissolution, using the rate from Sánchez et al. (2006), leads to analcime precipitation. Model results indicate that smectite dissolution in the presence of an OPC concrete liner in a repository in clay is about 4 times more relevant than in the case of the repository in granite. (PEBS Project Deliverable 3.5.4).

Magnetite precipitates near the canister. The thickness of the altered zone at the canister-bentonite interface in the presence of the high pH plume is 4 cm. This is half of the thickness of the altered zone for a repository in granite (PEBS Project Deliverable 3.5.4; Samper et al. 2014c).

The pH in the concrete increases due to portlandite dissolution. It also increases in the bentonite due to the penetration of the hyper alkaline plume from concrete. The precipitation of calcite, brucite and sepiolite buffers the hyper alkaline plume. The high pH plume extends throughout the bentonite and causes mineral precipitation, changes in the composition of the exchanged cations and a reduction of the porosity of the bentonite. The pH is about 11 after 100000 years and later decreases to 9.5. The hyper alkaline plume from the concrete penetrates 1 m into the clay host rock after 1 Ma (PEBS Project Deliverable 3.5.4; Samper et al. 2014c).

Numerical models predict pore clogging in a 4 cm thick zone near the canister, 5 cm in the concrete-bentonite interface and 5 cm in the clay-concrete interface. Changes in porosity caused by mineral dissolution precipitation are significant throughout the bentonite and in a narrow band (25 cm) of the host rock.

The numerical results of the UDC models of the interactions of concrete with bentonite and clays and the effects of the hyper alkaline plumes generated by concrete on engineered and geological barriers have been compared to the results reported by Shao et al. (2013), Kosakowski and Berner (2013) and Berner et al. (2013). Shao et al. (2013) performed reactive transport simulations of the Maqarin marl rock natural analogue for 500 years with a 1D model by using the OpenGeoSys-GEM code. Their results show pore clogging in the marl rock exposed to the hyperalkaline plume in a band of 5 to 10 mm due to the precipitation of ettringite and CSH phases. According to the UDC numerical model, the pores of the clay rock become clogged after 20,000 years. Kosakowski and Berner (2013) presented numerical reactive transport calculations to evaluate the geochemical evolution at cement/clay interfaces for different transport scenarios through several Swiss clay rocks for a time span of 2,000 years. Their calculations show that the effects of geochemical gradients between concrete and clay

materials are very similar for all investigated host rocks. The mineralogical changes at the material interfaces are restricted to narrow zones for all host rocks. The extent of strong pH increase in the host rocks is limited. The calculations show massive porosity changes due to precipitation/dissolution of mineral phases near the interface, in line with other reported transport calculations on cement/clay interactions. They concluded that the degradation of concrete materials will be limited to narrow zones. The porosity reduction will be restricted to very narrow zones. The clogging of the pore space leads to a strong reduction of diffusive fluxes across the interface, which essentially stops the geochemical alteration processes and significantly slows down mass transport across the interface. However, connecting the evaluated temporal porosity evolution to realistic and true times is very difficult. The calculations of UDC share most of the features of the results of Kosakowski and Berner (2013) such as narrow alteration zones with pore clogging. Berner et al. (2013) presented the simulations of the interactions of a MX-bentonite buffer with a low-pH concrete and the Opalinus Clay. Calculations were performed for 30000 years by using the OpenGeoSys-GEM code. Their results show that the thickness of the zone containing significant mineralogical alterations is at most a few tens of cm in both the bentonite and the Opalinus Clay adjacent to the liner. The precipitation of minerals causes a reduction in the porosity near the bentonite-concrete and concrete-Opalinus Clay interfaces. The effect is more pronounced and faster at the concrete liner-Opalinus Clay interface. The simulations reveal that significant pH-changes (i.e. pH >9) in the bentonite and the Opalinus Clay are limited to zones less than 10 cm thick after 30000 years. UDC calculations share also many of the results of Berner et al. (2013). The zone of the bentonite affected by clogging is 15 cm while clogging affects 10 cm of the clay formation. Similar to Berner et al. (2013), UDC results show that smectite dissolution is very small (less than 0.2 % after 1Ma). The model of Berner et al. (2013) predicts that the pH is around 8 after 100,000 years and the pH front in the clay formation reaches about 10 cm. The UDC model predicts a pH of 11 at $t = 100,000$ years and a pH of 8 after 1 M years. The front of high pH penetrates 14.5 cm after 1 Ma. This discrepancy in pH predictions is due to the fact that Berner et al. (2013) considered a low-pH concrete while UDC considered OPC.

4.4.5 Key Issues Representing Uncertainties related to Case 4, Identified during PEBS

4.4.5.1 Bentonite Alteration at Temperatures above 100 °C

(see also Section 4.2)

At temperatures at and beyond 100 °C and full saturation the redistribution of soluble minerals within the bentonite matrix might cement parts of the intergranular porosity of the bentonite, the swelling pressure of bentonite might be somewhat reduced; and with high activities of potassium some illitization might occur. High temperatures may also lead to a slight increase in CEC.

Under unsaturated conditions temperatures of 100 °C induce the formation of saline fronts moving towards the hotter zones, increasing the concentration of chloride and sulfate at the interface, which may be relevant to iron corrosion (Turrero et al. 2011, PEBS Deliverable D2.3.3-1: Laboratory tests at the interfaces - First results on the dismantling of tests FB3 and HB4). Concrete and iron interfaces act as sinks of soluble salts (Cl, S) coming from the clay formation porewater or migrated through the bentonite. A redistribution of exchangeable cations within the bentonite will take place: Increase of Mg and K in areas close to the interface (70 %) and of Na near the hydration. At 100 °C, Precipitation of Ca phases (gypsum-anhydrite) occurs on Fe powder. There is also evidence of thermally-induced mineralogical changes such as silica

precipitation at the interface (600 μm): Cristobalite beads (size around 100 nm) were found on Fe interface in cells dismantled after 54 and 82 months of operation. The formation of very small amounts of random interstratified phases in the vicinity of the interface due to long-term exposure to high temperature is also possible (New results in HB5 and FB4 cells, unpublished).

Despite the above observed phenomena, periods of temperatures up to around 100 °C do not seem to impede the bentonite to fulfil its safety functions, as these reactions are predominantly at the interfaces with the canister and rock, thus the majority of the barrier is unaffected by these processes.

4.4.5.2 Uncertainty in the Bentonite Hydration Rate

The visual evidence of the production of a bentonite cemented zone near a dissolving lime mortar (small cells) indicates a process of clogging that may reduce the rate of hydration. However, in the small cells experiments, the bentonite became fully hydrated. The consequence of this type of phenomenon in the hydration rate at long term has to be addressed.

4.4.5.3 Uncertainty in Chemical Reactions and Parameters for Modelling

Significant advances in geochemical modelling of the EBS have been achieved within the PEBS project. Some uncertainties have been addressed, evaluated and resolved. The uncertainties in $\text{H}_2(\text{g})$ gas generation, diffusion and transport, and the formation of gaseous phases were addressed within the framework of other Euratom Projects such as FORGE. Uncertainties in equilibrium constants and their dependence on temperature are judged to be insignificant.

A literature review was performed to evaluate the long-term relevance of the mechanical and chemical couplings induced by the corrosion products. The main conclusions of this review indicate that no expansion of the test cells was observed during the corrosion tests performed by Smart et al (2006a). The absence of the expansion was explained by the mechanical properties of the oxides. According to Smart et al. (2006b), natural analogues for anaerobic corrosion do not provide any experimental evidence for expansion caused by iron corrosion.

The remaining uncertainties include:

- The identification of the secondary mineral phases that may form. For the most part, the coupled THCM numerical models of the small and medium cells developed by UDC capture the main observed trends in mineral dissolution-precipitation. However, the model predictions for ettringite, CSH and brucite are less accurate. The precipitation of CASH phases was not accounted for in the models due to the lack of Al thermodynamic data. The nature of low crystal size C(A)SH and MSH at the bentonite/concrete interface is still unclear (Fernández et al. 2013).
- The kinetic data (rate laws, catalytic effects, rate constants, reactive surfaces)
- The identification of chloride absorbing phases at cement and iron oxides
- The impact of the low porosity layer at the cement boundaries on bentonite hydration (see section above) and on the spatial extent of the alkaline reaction plume

These uncertainties influence the mineralogical alteration thickness. Sensitivity runs were performed to evaluate the dependence of the thickness of the altered zone near the canister-bentonite interface to several key parameters and processes including the corrosion rate, the water flow through the granite/bentonite, the dependence of corrosion on environmental

conditions, the diffusion coefficient and the selectivities of the bentonite. The results show that this thickness is generally bounded within the predicted values of less than 10 to 12 cm.

4.4.5.4 Time-scale of Some Geochemical Reactions

Some geochemical reactions such as smectite dissolution are commonly disregarded in short to medium-term geochemical models (e.g. lab and in-situ tests) because they are regarded not relevant in the short term. However, the long-term relevance of such reactions has to be ascertained.

Savage et al. (2010b) studied the potential contribution of the smectite hydrolysis to the long-term geochemical processes in a KBS-3 bentonite buffer. They concluded that smectite dissolution may be significant for the future geochemical state of a buffer, but the time-scale of this process is too long for experimental verification. They also suggested the importance of further studies through field observation of natural clay-water systems that have evolved over long time scales.

UDC performed full geochemical calculations for repositories in granite and clay (Spanish Reference Concepts) using UDC codes by considering the kinetic dissolution of bentonite for one million years. Model simulations performed with and without kinetic smectite dissolution have been compared to test the relevance of this process. For a repository in granite the cumulative amount of smectite dissolution after 1 Ma is approximately = 0.25 mol/L. This amounts to a decrease of bentonite mass of less than 0.2 % after 1 Ma. The decrease in porosity of about 0.01 is compensated with the amount of precipitated analcime. Smectite dissolution has the beneficial effect of leading to a slight decrease (1 cm) of the thickness of the altered bentonite zone (PEBS Project Deliverable 3.5.4; Samper et al. 2014c). Model results indicate that smectite dissolution in the presence of a concrete liner is about 4 times more relevant than in the case of the repository in granite (PEBS Project Deliverable 3.5.4; Samper et al. 2014c).

As a conclusion, the long-term effect of the kinetic dissolution of smectite on bentonite porosity and on the thickness of altered bentonite is small for the EBS of repositories in granite and clay according to the Spanish reference concept.

4.4.6 Evaluation of Uncertainties - Remaining Issues

4.4.6.1 Degree of Alteration, Properties and Thicknesses of the Altered Zones

From a scientific perspective, the most relevant chemical processes involved in the interactions of corrosion products, bentonite and concrete have been identified, especially once the bentonite is fully saturated. For unsaturated conditions, the interactions are more complex. However, most of the corrosion will take place under saturated anoxic conditions.

Several uncertainties were resolved or proven insignificant by the PEBS investigations:

1. The thermal transient does not have a significant effect on the overall geochemical evolution of the EBS.
2. Corrosion products have no traceable influence on the mechanical state of the buffer.
3. The effect of smectite dissolution on bentonite porosity and on the thickness of altered bentonite is negligible.

As a result of UDC's modelling in PEBS as well as other comparable studies, thicknesses of altered zones will generally be bounded within values of less than 10 to 12 cm.

Modelling chemical reactions, especially the dissolution and precipitation of mineral phases, requires knowledge of the nature of the reaction (thermodynamic equilibrium or kinetic control), the equilibrium constants and kinetic rate parameters (rate laws, rate constants and reactive surface areas). Available data from laboratory experiments are not enough to provide well-defined geochemical systems. Therefore, there are uncertainties some of which have been addressed and evaluated for the long-term simulations.

The remaining model uncertainties include:

- The precipitation of secondary mineral phases at the concrete-bentonite interface such as CSH, CASH, zeolites, and iron silicates
- The precipitation of the secondary mineral phases at the canister-bentonite interface such as Fe-phyllsilicates and the reduction of Fe in the bentonite
- The rate laws, catalytic effects, rate constants and reactive surfaces of kinetic mineral dissolution/precipitation
- The clogging of the bentonite and clay pores. The precipitation of the minerals could form a low-porosity layer which could affect the hydration rate and the subsequent geochemical evolution of the system (Cuevas et al. 2014). Their nature is still unclear.

Similar to Case 1, further research is needed to improve the knowledge of the microstructure of the bentonite and the role of the adsorbed interlayered water. The use of dual porosity models and the update of the porosity due to chemical reactions could help resolve some of the uncertainties identified in the lab experiments.

4.4.6.2 Effect of Alteration on Overall Buffer Performance

The altered zone in the bentonite near the concrete exhibits altered mechanical and hydraulic properties. The alteration at the concrete-bentonite interface and on the iron-bentonite interface will have an effect on swelling pressure which, for now, cannot be quantified. As long as the altered zones are thin compared to the unchanged bentonite portion, it can be expected that the overall swelling pressure will not be critically impaired. From the modelling an upper boundary for the thickness of the altered zones is predicted; it is, however, important to further reduce the uncertainty in this prediction.

With regard to the effect of alteration on the hydraulic performance of buffer, one result of experiments and modelling is the pore clogging in the altered zones, which would in itself mean an improvement by permeability reduction, provided that a sufficient overall swelling pressure of the buffer is reached. The potential effect of pore clogging on the hydration rate could, on the other hand, mean a longer duration of the unsaturated state of the buffer with more complex conditions.

5 Impact of the Improved Understanding on the Safety Functions

5.1 Introduction

In this chapter the link is made between the detailed process understanding as described in the cases in the previous chapter including the remaining uncertainties and the repository concepts and their associated safety functions with respect to the repository evolution in the transient stage.

Detailed process understanding of the behaviour of bentonite barriers is applied in performance assessment in a manner determined by the impact of the processes on safety functions of the EBS and host rock. The significance of the various processes depends on the disposal system design including hydraulic transport boundary conditions. Generic conclusions that apply to all disposal systems are thus difficult to make. This is presented graphically in Fig. 5.1. As a result the findings of the PEBS project are discussed below in the context of their application to specific disposal systems. These summaries draw on information presented in D1.1, which provides detailed discussions of the disposal systems, with particular reference to bentonite barriers that have been proposed in Sweden, France, Spain, Switzerland and Germany. The information on each of the disposal systems is only briefly reiterated in order to provide context for discussing the relevant project conclusions.

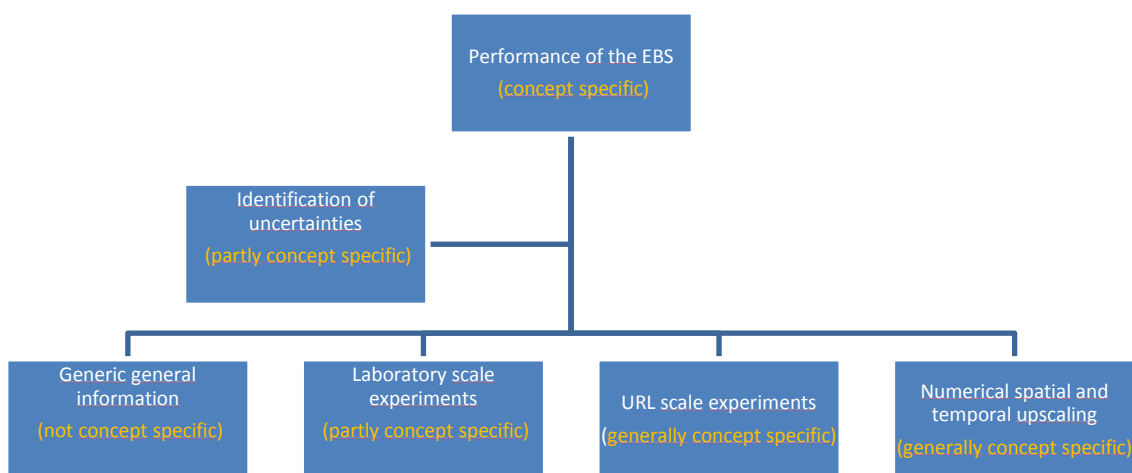


Fig. 5.1: Generic and concept specific information related to the performance of the EBS

Based on this integration conducted for the relevant concepts in the PEBS project an improved and more complete approach to integrating the thermal and re-saturation phase of the repository with the long term steady state phase is proposed. In this context the remaining significant safety-relevant uncertainties can be identified and the links with optimisation of the design can be established.

5.2 PEBS Project Results in Relation to the Nagra Disposal Concept (Switzerland)

The significance of findings from the PEBS project regarding the THMC evolution of the bentonite barrier in relation to its safety-relevant properties as well as the overall impacts on safety of the Nagra disposal system are discussed here. An overview of the evolution of the disposal system over time, with a focus on bentonite barrier performance, was given in D1.1. This is briefly reiterated here to allow the PEBS findings to be placed in context.

The repository concept in Opalinus Clay envisions an array of long (~ 800 m) parallel tunnels at a depth of 600 to 900 m containing SF or HLW canisters, with the region around the canisters filled with bentonite, as shown in Fig. 5.1a (Nagra 2002). The canisters would be constructed from thick-walled (12-14 cm) carbon steel (an alternative is copper-coated steel). The bentonite backfill is a combination of blocks beneath the canister and granular bentonite and has a reference saturated density of about 1.9 Mg m^{-3} . Because of the low hydraulic conductivity of the Opalinus Clay, the transport conditions after saturation of the backfill are diffusion-controlled. For tunnel support, a low-pH shotcrete liner would be used, with a hydraulic seal placed between every tenth canister (Nagra 2010). The hydraulic seal would comprise bentonite of higher density than that used around the disposal canisters, constructed using a combination of blocks and pellets. In the seal zone, 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 continuous hydraulic flow through the more permeable liner, as indicated Fig. 5.1b.

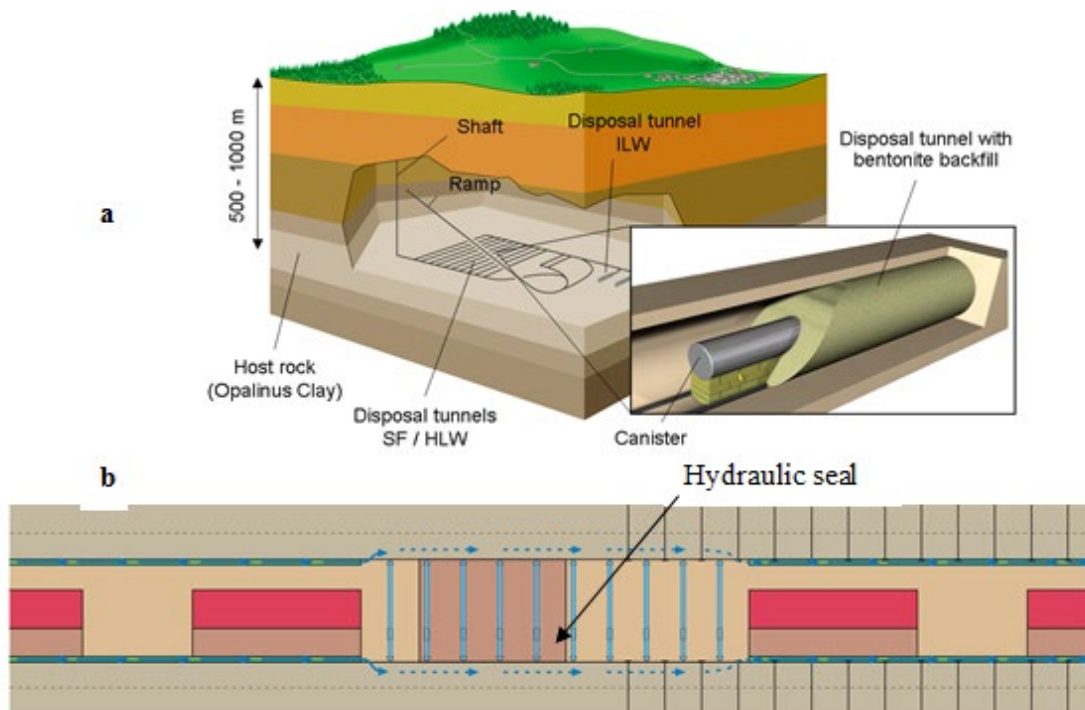


Fig. 5.1: Nagra disposal concept for SF and HLW: (a) repository layout and detail of EBS; (b) longitudinal section of emplacement tunnel showing liner and hydraulic seal

The performance requirements for the bentonite barrier are discussed in D1.1 (Tab. 4) ; the most important for overall performance are hydraulic conductivity of less than $10^{-11} \text{ m s}^{-1}$ and a swelling pressure of $>0.6 \text{ MPa}$. The requirements apply to the saturated state of the bentonite; in the short-term period prior to saturation the bentonite is required only to provide support to the excavation (limit further convergence), maintain the position of the canister and conduct heat to the rock.

The PEBS project has focused in particular on the behaviour of the bentonite in the short-term unsaturated condition, the evolution of properties during the transition to the saturated state and how the associated processes affect the achievement of long-term safety requirements. The following discussion focuses on the overall findings of the PEBS project and how they relate to the EBS performance in the Nagra disposal concept.

Case 1

In a repository for SF/HLW in Opalinus Clay, the buffer saturation process is controlled principally by the low permeability of the rock and homogeneous inflow conditions at the outer boundary of the bentonite are expected. The results from Case 1 show that when bentonite is wetted uniformly from the outside, a circumferential outer ring of saturated buffer with significant swelling capacity will develop.

Case 2

The HE-E experiment (see Section 4.2) provided a first large-scale test of the understanding of thermo-hydraulic behaviour of the Nagra near-field concept. The TH models that simulate the experiment capture the observed behaviour reasonably well. The temperature calculations show good agreement, while the initial degree of saturation within the bentonite barrier close to the canister after achievement of thermal steady-state is somewhat over predicted. An interesting observation from the results is that the degree of saturation in buffer close to the canister is extremely low (10 - 20 %). This is the case for both the blocks beneath the canister and the granular bentonite used as backfill elsewhere, despite the fact that the blocks were emplaced with significantly higher water content than the granular material. This illustrates the rapid redistribution of water content according to the thermal gradient. Temperatures in the buffer and in the rock near field ($<2 \text{ m}$) are reasonably well reproduced by the models. The size and the trend of the thermally induced hydraulic overpressures (in the range of 1 MPa) at a distance of several meters into the rock are also reasonably well reproduced. THM modelling results and their partial validation in the PEBS project lead to increased confidence in this type of modelling in terms of providing secure bounds for the range of impacts (under the assumption that no THM processes appear in the later stages that not have been observed in the early stages and that the boundary conditions do not change over time). However, as noted below, a full understanding of the impact of the early evolution period on the buffer can only be achieved by a combined suite of models and laboratory studies, the results of which are summarized below.

Based on the results from the HE-E experiment, TH models appear to describe T and RH distributions at the time that maximum canister and buffer temperatures are reached. Their capacity to represent the system at later times will depend to some degree on the buffer re-saturation rate, which is predominantly determined by the hydraulic pressure field in the host rock. The time-dependent temperature evolution for the reference re-saturation rate is shown in Fig. 5.2. Based on the sensitivity analyses performed, a re-saturation time between 50 - 100 years is estimated whereby reasonable variations in the host rock permeability have a stronger impact than variations in the bentonite permeability. The time window for the

temperature in fully water-saturated bentonite buffer to exceed 100°C (with a maximum of 115 °C) ranges from 50 to 180 years. Such TH models require further testing, which is expected to be achieved through continuation of the HE-E experiment and initiation of the full-scale FE experiment.

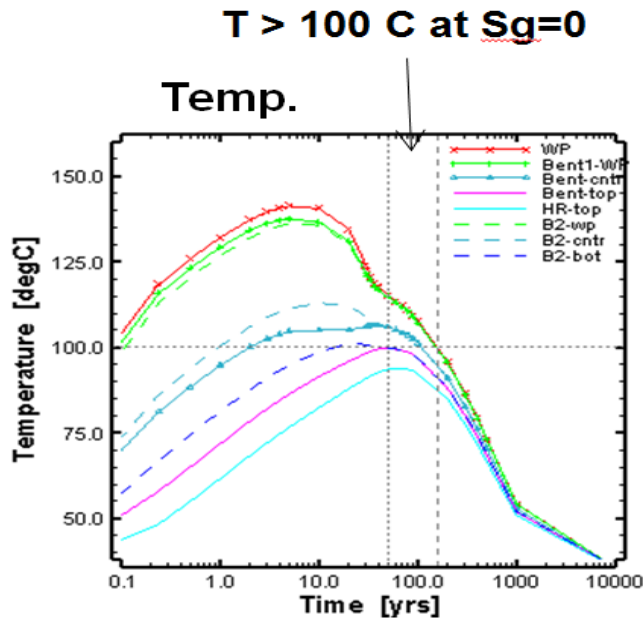


Fig. 5.2: Simulation of the temperature evolution in the near field (reference WP3.5 report)

In the HE-E experiment it has been shown for the first time that a very dry zone will immediately develop near the canister. There is need to continue this experiment to establish the impact of this period on later stage hydraulic and swelling properties. In the meantime, laboratory and field studies continue to provide useful insights into these questions. A minor reduction in swelling pressure may occur as a result of exposure of bentonite to unsaturated conditions at temperatures exceeding 100 °C. The results are based on a relatively sparse dataset (especially for pellets) and more systematic characterisation work is needed in the future.

Studies of the hydraulic and swelling properties of saturated bentonite heated to temperatures of up to 150 °C in laboratory and field experiments for periods of months to several years show negligible changes in hydraulic conductivity and swelling pressure, but some increase in shear strength. Nonetheless, there is clear evidence that reasonable swelling pressure and low hydraulic conductivity are likely to be maintained, i.e. minimal requirements of a hydraulic conductivity of less than 10^{-11} m s⁻¹ and a swelling pressure of >0.6 MPa are expected to be achieved. The overall changes to safety-relevant properties do not appear to be significant.

Case 3

In the Nagra concept, there are significant differences between the density of blocks below the canister (initial dry density ~ 1.7 Mg/m³) and granular bentonite used to backfill after canister emplacement (initial dry density 1.45 Mg/m³). The EB experiment results show that the denser blocks below the canister will swell and slightly displace the canister upwards, thus somewhat

compacting the granular bentonite. Thus some degree of homogenisation of the bentonite will take place during re-saturation. The EB experiment also shows that all voids are effectively filled by the swelling bentonite. Models have indicated that the final heterogeneity of mass in a bentonite barrier may depend on the wetting history.

Case 4

Considerable progress has been made in modelling long-term THMC effects on the bentonite barrier, in particular in gaining insights into reactions at steel canister/bentonite and concrete liner/bentonite interfaces. Nonetheless, many uncertainties exist in these models and it is sometimes only possible to establish the limiting effects of these reactions through a combination of mass transport and mass balance considerations. Relevant findings from the PEBS project for the Nagra safety assessment perspective are summarized below.

Short-term laboratory studies show that the montmorillonite content is unaffected by heating to up to 150 °C under both unsaturated and saturated conditions. Application of geochemical models of smectite to illite conversion to the specific time-temperature evolution of the bentonite backfill in the Nagra concept suggest that only minor transformation is likely to occur, even over hundreds of thousands of years. The main reason for this is the relatively short period of time of exposure to elevated temperatures.

Although the temperatures in the region close to a canister may be in the range of 130 - 150 °C for some decades, the impact of a short period of time with such a low degree of saturation is expected to be very limited. Chemical reactions are suppressed (e.g. corrosion is negligible below about 40 % RH (reference) and smectite alteration is slowed. Experiments of up to 6 years on the interaction of bentonite with iron corrosion products under reducing conditions indicate no new iron silicates are formed and a penetration depth of Fe into the bentonite of only a few mm. Reliable long-term modelling of the process has not been achieved because of a large number of uncertainties associated with kinetics of reactions, porosity changes and thermodynamic data for Fe-silicates. Indications from models are that the interaction zone may be 1-10 cm after some tens of thousands of years (Bradbury et al. in press). In the case of an OPC concrete liner, the results of long-term modelling of the interface suggest a quite limited alteration zone in the bentonite. However, the uncertainties in the coupled HC models appear to be significant, thus Nagra proposed the use of a low pH concrete liner rather than OPC to further limit the extent of alteration. For this material, even the case of a conservative mass balance-based model suggests a maximum of only 4 cm of bentonite would be altered (Savage 2013).

Swelling pressure reduction that arises from hydro-chemical alteration is likely to occur over many tens of thousands of years, as a result of the slow dissolution and alteration processes at the canister/buffer and liner/buffer interfaces. Convergence of the host rock may also occur over the long term (the in situ stress is about 15 MPa at 600 m), which could lead to some compaction of the bentonite. The rate of convergence is unclear and remains to be determined in modelling and experimental studies. The minimal requirements for the bentonite backfill of a hydraulic conductivity of less than 10^{-11} m s⁻¹ and a swelling pressure of >0.6 MPa are expected to be achieved.

It is noted that sensitivity analysis calculations of radionuclide transport and radiological consequences have been performed for cases in which significant reductions in safety-relevant properties of the bentonite have been assumed and the results show very small consequences (Nagra 2010).

5.3 PEBS Project Results in Relation to the Swedish Disposal Concept (Sweden)

The main function of the clay buffer in the Swedish KBS-3 concept (Fig. 2.1) is to restrict water flow around the canister. This is achieved by choosing a buffer material with a low hydraulic conductivity after water saturation. This makes diffusion the dominant transport mechanism. The material should also have a sufficient swelling pressure, making the buffer self-sealing. The clay material's montmorillonite content is a key property for the safety functions of the buffer.

The main aim of the project PEBS has been to evaluate the sealing and barrier performance of the EBS with time, through development of a comprehensive approach involving experiments, model development and consideration of the potential impacts on long-term safety functions. The results from PEBS will have an impact the description of the reference evolution of the future assessment. The impact of the different cases is described in the following text.

Case 1

In the SR-Site assessment (SKB 2011) the saturation times for both backfill and buffer range from a few tens of years to several thousand years. Examples from across this entire range are likely to arise at Forsmark, since rock properties (matrix hydraulic conductivity and presence and characteristics of fractures) are the primary controls, with backfill and buffer properties only a secondary consideration. While the saturation time only has indirect effect on the safety functions, the Swedish regulator (SSM) still considered the assessment in SR-Site incomplete and requested additional information. The view of SSM was that conceptual uncertainties associated with thermal, hydraulic and mechanical processes in the near field of a repository has been investigated since many years, both theoretically and through large-scale experiments, in a number of EU projects. This information has, according to the judgment by SSM, not been optimally utilized in the calculations of the re-saturation phase in SR-Site. SKB should analyse the relevance of the conclusions from other extensive experiments, such as FEBEX, and include a systematic analysis of conceptual uncertainties which can influence the modelling of the evolution of the re-saturation. It has for instance been suggested that the influence of thermo-osmotic effects and the occurrence of a threshold gradient has a significant influence in particular for those longer re-saturation times which occur in SKB's case. As described in section 4.1.2.4 this has been extensively studied within the PEBS project and the results were of considerable value for SKB in the preparation of requested information to SSM.

Case 2

Case 2 is of less relevance to SKB, since the maximum temperature is set to 100 °C. Still, the study is still importance since:

- Data for temperatures >100 °C, will improve the overall process understanding and the capabilities of the THM-models.
- A vapour phase will most likely be present in the buffer for a significant time, even if the temperature is <100 °C.

The conclusions from PEBS that the THM-formulations can be used and that no significant alteration of the HM-properties can be observed for temperatures above 100 °C strengthen the argumentation in the safety case.

Case 3

The conclusion from SR-Site assessment (SKB 2011) was that if the buffer and backfill is installed as envisaged by the reference design, the buffer density and swelling pressure will homogenise to a situation where the relevant safety functions will be upheld. Modelling of the large-scale tests and comparison with measurements confirm that the material model of unsaturated bentonite blocks and the calculation technique used are relevant for modelling the homogenisation process. The uncertainties are mainly the material models, which are very complicated, and the parameter values. Although they have been verified for the one-dimensional case of swelling and homogenisation of the bentonite rings and pellets between the canister and the rock, the two-dimensional case involves more degrees of freedom for the variables and more interactions like the friction between the bentonite and the rock or canister.

The results from PEBS confirmed that the homogenization of a bentonite buffer is efficient; even it is installed as a mixture of high density blocks and low density pellets. The EB experiment has confirmed that a highly saturated bentonite barrier was able to seal all the initial voids. This observation is important since average dry density in EB was rather low and the FEBEX-bentonite is mainly in a calcium form, which should give a low swelling pressure at low density. The homogenisation in EB stands in sharp contrast to the observations from the tunnel backfill in the prototype repository in S ap o. There, the backfill consisted of a bentonite/crushed rock mixture and the resulting homogenisation was very poor.

The results from FEBEX mock up shows that the buffer achieves the required performance over time as saturation increases, as shown by the sufficient and homogeneous swelling pressures developed in the highly saturated outer layer of the buffer. This observation is important since it confirms that the seal between the buffer and rock will develop long before full saturation is reached.

Case 4

The results from the geochemical work in PEBS are less conclusive. They do however confirm observations from earlier studies:

- No newly-formed iron-rich clay phases of the bentonite were detected in experiments lasting two years
- No alteration of montmorillonite is observed in the short time frame (2y)
- Geochemical modelling involves significant challenges
- Bentonite-cement interaction is restricted to a limited interface zone
- The lack of iron-rich clay phases is consistent with the findings from the ABM project (Svensson et al 2011).

5.4 PEBS Project Results in Relation to the French Disposal Concept

In France, the concept for vitrified waste is based on micro tunnel (0.7 m in diameter, minimum length of 40 m, Fig 5.3a/b) with a steel casing to emplace and retrieve easily the canister during the lifetime of the casing. No bentonite is in contact with waste canister, but bentonite seals are placed at different locations in access drifts after emplacing the waste. That means that bentonite is not exposed to significant heat (Fig 5.3c) in the French concept (compared to the other concepts presented in this report). Other local thermal processes are very limited in time and space, such as hydration heat produced during strengthening of the low pH concrete support

walls or thermal rebalancing in the near field Callovian-Oxfordian. Thus the seals nearby the HLW tunnels experience a temperature rise of a maximum of 20 °C to 25 °C. The temperatures of these seals are therefore between 30 °C and 50 °C for up to several thousand years. The temperature rise and cooling kinetics are slow, of the order of one degree Celsius in a few decades.

The knowledge on swelling clay-based materials, including the feedback from many tests and models up to multi-metric scales from international programs, indicate that a permeability lower than 10^{-9} m/s can be achieved with different types of swelling clay (pure or mixed), corresponding to a wide range of dry densities and different methods of shaping and installation. ANDRA fixed the homogenized water-saturated permeability of the swelling clay core, including its interfaces with other materials, in the order of 10^{-11} m/s. In addition to its hydraulic performance, mechanical performance is also aimed for the swelling clay-based core. By the development of a swelling pressure with saturation, the clay core may indeed limit the deformations of Callovo Oxfordian claystone with which it is in contact and thus limit the possibility of reactivating of the damaged zone formed during the excavation and emplacement of the seal. For a swelling pressure of less than the effective in situ stress, deferred deformations of the argillites could lead to compaction of the core with a competition between creep and increased swelling pressure leading to an “unloading” of the damaged zone until equilibrium is reached. Note that, conversely, excessive swelling pressure beyond the natural effective stress would likely lead to new mechanical disorder in the damaged zone. Therefore, the definition of the requirements for a swelling clay material core and its mode of implementation are guided by the search for a favourable swelling pressure taking all phenomenological elements above into account as well as technical constraints associated with industrial use. This leads at this stage to a target swelling pressure of the core around a few megapascals.

Due to the location of seals inside the storage zone, especially in terms of distance from potential sources of chemical interactions or processes that influence the chemical processes (cf. heat load above), their chemical evolution is generally local and develops mainly from the interfaces between different materials. Qualitatively, the experimental data and simulations indicate that the cementitious interactions (coupled with degradation products of the packages for some IL-LL waste cells) have limited extensions (in the order of several decimetres for remineralised areas and one meter for the front of high pH). Given the large dimensions of the seals and the use of low pH concrete for portions of the liner and support walls, sources of chemical interactions of adjacent structures (all seals) and those of useful parts of cells for IL-LL waste (IL-LL cell seals only) are highly buffered on both sides of the seals.

Note that the re-saturation of the swelling clay core of the storage level seals will mainly occur by water that has percolated through the portions of the maintained liner and the support walls that are both built with low pH concrete. For a low initial degree of water saturation of the order of a few percent (the case of bentonite pellets), the chemical degradation of the core, mainly occurring by dilution of cementitious water and chemical reactivity in the host rock clay, should impact at most approximately 10 % of the total amount of the swelling minerals (smectites). It can be therefore reasonably considered a chemical evolution of seals located mainly at the interfaces and with limited extension. PEBS results confirm this trend.

The evolution of bentonite barrier performance (seals) over time remains an issue for safety calculations, and findings from the PEBS project regarding the THMC evolution of the bentonite barrier give insight to the safety-relevant properties as well as the overall impacts on safety. After the seal is emplaced, its phenomenological evolution can be decomposed into two main phases:

- The transitional period of re-saturation to achieve total water saturation of the seal and of the full storage area
- The long-term period after total saturation

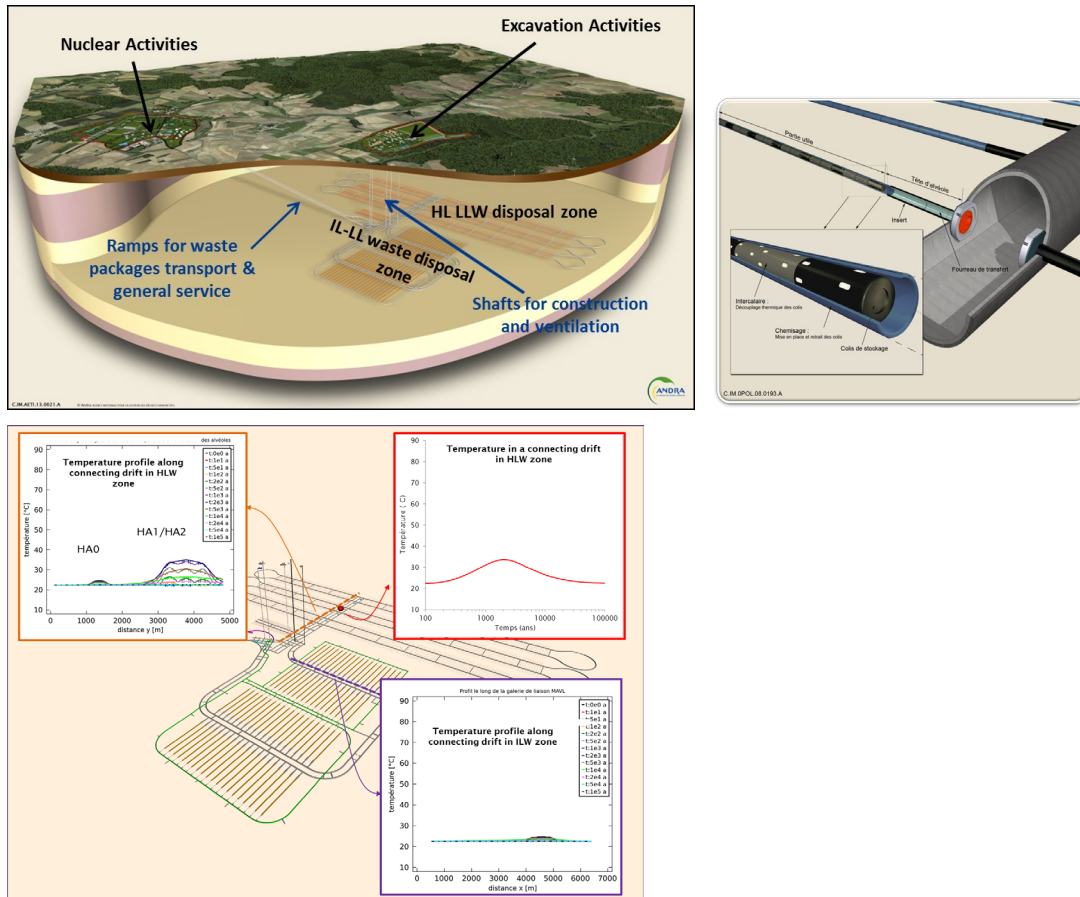


Fig. 5.3: NDRA disposal concept for ILW and HLW: (a) repository layout; (b) HLW cell concept: horizontal micro tunnel with steel casing; (c) Level of temperature in different drifts

The dismantling of the EB experiment, the visual observations and the on-site and laboratory tests results have confirmed that the bentonite barrier was highly saturated, sealing all the initial voids. Also, the hydraulic conductivity was as low as foreseen. However, even though some degree of homogenization of the bentonite materials has already taken place (the blocks have decreased in density and the GBM has increased in density), there is still a trend for the moisture content to increase towards the bottom of the section (while the dry densities are lower than in the upper part). It is worth noting when analysing these results that, in this specific experiment, a noticeable initial segregation took place due to the GBM emplacement procedure used. Given the large cross-sections of the excavations (up to 100 m² in the galleries or in the ramps) in the repository, potentially irregular surfaces of their walls and technological difficulty in ensuring a uniform filling of the entire volume emplacement technique have to be checked to handle this segregation problem and to reduce it. Within the EU DOPAS project, ANDRA's technological tests program should contribute to the strengthening of the technical capacity of

realization of the seals and to the definition of their initial state (FSS implementation test of a full-scale seal and test of liner removal in the Bure URL). But EB dismantling has confirmed that the bentonite barrier was highly saturated and the bentonite blocks and GBM, which initially had significantly different dry densities, had developed rather similar average densities and that even if emplaced with relatively low average dry density (1.33 t/m^3 in this case) the permeability tests performed confirm that, after saturation, the barrier had a low hydraulic conductivity (k_w) equal to or lower than $5 \times 10^{-12} \text{ m/s}$. There is clear evidence that reasonable swelling pressure and low hydraulic conductivity are likely to be maintained even if the EBS endures high temperature at early stage. The overall changes to safety-relevant properties do not appear to be significant.

THM modelling results presented in the PEBS project shows new progress and increases confidence in this type of modelling approach. However long-term deformation of the Callovo Oxfordian claystone is known, although evolution of rates of deformation in the long term remain uncertain and the “equilibrium” between convergence versus swelling of the core at full saturation remain to better understood. Due to the size of the drift, bentonite pellets are considered as the more industrial technique for emplacement (see EU DOPAS project) but bigger pellets (compared to those used in the PEBS project) have to be used and models developed in PEBS have to be checked in this case to see their ability to predict the core behaviour during saturation.

5.5 PEBS Project Results in Relation to the Thinking on Disposal Concepts in Germany (GRS)

In Germany, no decision has been made so far about the site of a future final repository for heat-producing radioactive waste. Not even the host rock type has been fixed. Clay is one of the options, but two different clay types are under discussion: the Opalinus clay in Southern Germany or the Barrême/Hauterive clay in Northern Germany. So far, only rough preliminary concepts for a repository in clay have been developed. One option is a single-level mine with two shafts, a network of service and charging drifts and two fields of emplacement boreholes, one for Spent Fuel elements and one for vitrified waste from re-processing. Specific seals are emplaced on top of the boreholes, at the ends of the drifts and in the shafts. The seals are composed of bentonite and concrete elements and should have a durability of at least 50 000 years. Sketches of the structure of this concept and an emplacement borehole as developed in the framework of the German national project ANSICHT are presented in Fig. 5.4 (Lommerzheim 2012).

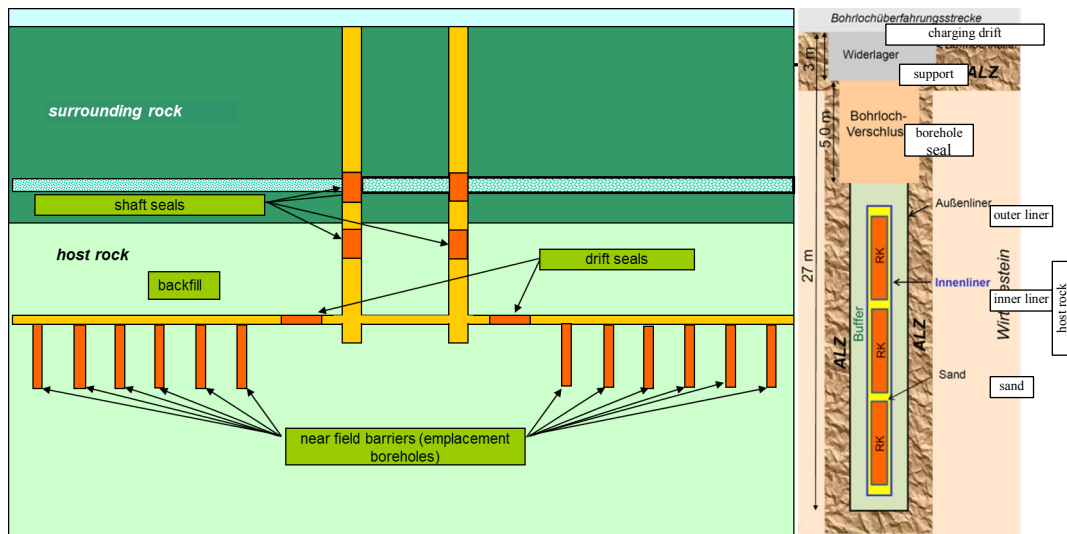


Fig. 5.4: Preliminary concept for a German repository in clay (by DBE)

The German system for proof of long-term safety is based on the idea of safe isolation of the waste. That means it has to be proven that the wastes will remain confined within a limited region of the host rock, called the isolating rock zone (IRZ), and that insignificant amounts of radionuclides can escape from the IRZ. The corresponding safety function is called containment and can be seen as the basic safety function a future repository has to provide (BMU 2010).

For a repository in clay, an effective limitation of contaminant transport is based on three essential retention mechanisms:

- The spread of radionuclides by advective transport should be, at best, comparable to that caused by inevitable diffusion. A low permeability of the host rock and the seals makes sure that the advective transport of radionuclides is inhibited or at least essentially restrained.
- Radionuclide diffusion should be as low as possible. Actinides typically show high sorption on clay materials, which effectively reduces the diffusion of the most relevant radionuclides. Additionally, the solubility of most contaminants is limited, so that the boundary concentration for the diffusion process is low.
- The host rock is inevitably disturbed by waste emplacement, which might impair its good isolation properties. The clay should recover as soon as possible from such disturbances. The envisaged clay formations have a good recovery potential, which should make sure that the isolation capacity is preserved throughout the entire reference period.

To fulfil the containment requirement, a repository in clay should be designed with specific regard to these properties.

Uncertainties that have the potential to influence one of the mentioned retention effects or even to impair their efficacy are most relevant from the German point of view. The work in PEBS has contributed to identifying and reducing such uncertainties.

Case 1:**Uncertainty in Water Uptake in the Buffer below 100 °C**

The process of water uptake of the clay buffer is of lesser relevance for the containment safety function, since fluid movement is not directed outwards and cannot cause contaminant release. The effects investigated can have an influence to the time when a possible contaminant transport starts, but since the containment has to be made sure anyway, the re-saturation time is not expected to impair this safety function. It was also stated by the investigators that the time to reach full saturation is not a critical parameter in the safety assessment. Therefore, the findings of Case 1 are assumed to be of minor interest with regard to the German safety concept.

Case 2:**Uncertainty in the Thermal Evolution of the Buffer above 100 °C**

The time phase with temperatures above 100 °C could become relevant for the containment safety function if it led to permanent alteration of the bentonite properties. Mineralogical changes of the buffer material might increase its permeability, reduce its sorption capacity or impair its ability for recovery. So all three retention mechanisms might be affected and the containment capability could be weakened. Such effects occurring at high temperatures have been addressed within PEBS, which has contributed to identifying the relevant uncertainties and focusing the attention to the relevant processes. It was found, for example, that the main physical parameters can be expected to remain stable at temperatures above 100 °C, but a limited effect on swelling pressure is possible.

Case 3:**Uncertainty in HM Evolution**

The long-term properties of the bentonite are essential for the buffer to provide the required containment capability. Uncertainties in processes influencing these long-term properties were specifically addressed in PEBS. Density gradients occurring in the buffer during the hydration phase could become relevant if they remain existent in the long-term. It was found that under certain circumstances, depending, for example, on the water uptake properties of the bentonite, the permeability of the buffer can become inhomogeneous. This might directly affect the above-mentioned requirement of low permeability and is an important uncertainty to be taken into account in the assessment of the containment capability.

Case 4:**Uncertainties in Geochemical Evolution**

Geochemical effects might have the potential to permanently alter parts of the EBS and to impair its retention capability. For this reason, the uncertainties related to such effects should be taken very seriously. With regard to the three retention mechanisms mentioned above, the following questions concerning geochemical effects that are likely to occur in the repository are of specific interest:

- Can the permeability of the buffer or the seal materials be increased?
- Can the sorption of actinides be significantly reduced?
- Can the self-healing properties of the buffer and seal materials be impaired?

The investigations undertaken in PEBS are a valuable contribution to identifying and reducing uncertainties about these questions. Bentonite alteration seems to be limited to a layer of several millimetres up to a few centimetres from the canister/bentonite or the concrete/bentonite interface, even over the long-term. This result might contribute to increasing the confidence in the long-term stability of the buffer and the seals.

With regard to the German safety concept the investigations conducted in PEBS provide a number of interesting results, specifically addressing uncertainties in buffer evolution triggered by thermal, hydro mechanical and chemical effects. A detailed analysis of all relevant results with respect to increasing the confidence in a long-term prognosis about the containment capability of a specific repository site in clay cannot be given here, because the current concept is generic and was designed without any knowledge about the actual geological conditions.

5.5 PEBS Project Results in Relation to the ENRESA Disposal Concept

The significance of findings from the PEBS project regarding the THMC evolution of the bentonite barrier in relation to its safety-relevant properties as well as the overall impacts on safety of the ENRESA disposal system are discussed here. An overview of the evolution of the disposal system over time, with a focus on bentonite barrier performance, was given in D1.1. This is briefly reiterated here to allow the PEBS findings to be placed in context.

The repository concept in granite is based on the disposal of spent fuel in carbon steel canisters in long horizontal disposal drifts. Canisters are surrounded by high-density bentonite. The canister measures 4.54 m in length and 0.90 m in diameter, and contains 4 PWR or 12 BWR fuel elements in a subcritical configuration. The thickness of the wall of the canister is 0.10 m at the cylindrical wall and 0.12 m at the ends, and is capable of withstanding the pressures to which it is subjected under disposal conditions and of providing a minimum period of containment of one thousand years. After being unloaded from the reactor, the fuel elements are temporarily stored for their thermal power to decay to a level at which they may be disposed of with a total thermal power of 1,220 W per canister. A total 3,600 canisters will be required for the final waste inventory of spent fuel estimated for the Spanish nuclear power programme.

Canisters are disposed in cylindrical disposal cells, constructed with blocks of precompacted bentonite. Pre-compacted bentonite blocks, of 1,700 kg/m³ dry density (in order to achieve a final dry density of 1,600 kg/m³), are used. The blocks are initially non-saturated (degree of saturation of 66 %). The disposal drifts of 500 m in length and 2.4 m in diameter (see Fig. 5.5) are located at a depth of 500 m in the host formation. The separation between canisters is determined mainly by thermal constraints. Separations of 2.0 m between canisters and 35 m between disposal drifts have been established, in order not to exceed a temperature of 100 °C in the bentonite.

DISPOSAL CONCEPT

- **Deep disposal**
- **Crystalline rock**
- **Spent fuel**
- **Carbon steel canister**
- **Horizontal emplacement**
- **Bentonite buffer**

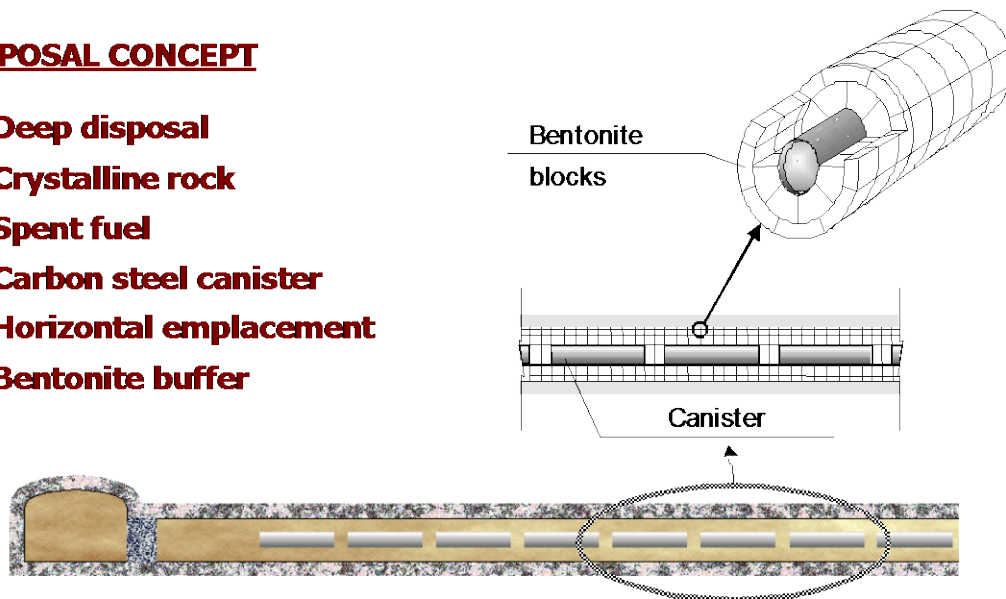


Fig. 5.5: Longitudinal section of a disposal drift

Once a disposal drift is completed, it is sealed with a 6 m long seal made of bentonite blocks and closed with a concrete plug at its entry. After completion of all the disposal drifts, main drifts, ramp, shafts and other remaining rock cavities will be backfilled with a mixture of bentonite and natural sand or an appropriate crushed material. The backfilling material will consist of 10 % bentonite (increasing up to 20 % at the top of the drifts) and suitably graded sand.

The bentonite buffer is required to maintain a large diversity of safety functions (D1.1), which can only be fulfilled once the bentonite saturates and swells, closing tightly the construction gaps between the bentonite blocks and the drift wall or the canister wall on the one hand and between the blocks themselves on the other. The gaps will close quickly upon contact with groundwater; in the case of the outer gaps, this will happen shortly (weeks or months) after buffer emplacement. Nevertheless there are not safety functional requirements applicable during the time the canister provides absolute containment. During the re-saturation of the buffer the main concern is the preservation of the favourable properties of the buffer material. As the safety functions assured by the buffer are accounted for the full duration of the quantitative safety assessment (in the scale of the million years) its properties have to be preserved at a sufficient level for commensurable periods of time.

The following discussion focuses on the overall findings of the PEBS project and their potential impact on ENRESA safety assessment for granitic rock.

Case 1:

Uncertainty in Water Uptake in the Buffer below 100 °C

The uncertainties and modelling discrepancies that were identified in ENRESA (2000) and reported in D1.1, after in depth analysis in PEBS, are now only recognized in the later phase of saturation, when the barrier already fulfils its safety functions. It is now possible to state that conservative values of the safety indicators (such as swelling capacity; hydraulic conductivity;

and thermal conductivity) related to the bentonite saturation can be predicted with reliability. The context, from a long-term safety perspective, has been clearly improved; and in any case, the residual uncertainty in water uptake during the later phase of saturation might even be qualified as conservative: it is likely that advective flow conditions from the host rock towards the buffer will be kept longer than estimated by the models.

Case 2:

Uncertainty in the Thermal Evolution of the Buffer above 100 °C

In the Spanish concept in granitic rock, there exists a safety function related to the temperature limit (<100 °C) in the bentonite in order to avoid mineralogical transformations that could result in the degradation of some bentonite favourable properties. The findings of this case are therefore of no interest to the current Spanish concept.

Case 3:

Uncertainty in HM Evolution

Uncertainties are essentially related to the time evolution of the density gradients generated in the buffer during the hydration phase that could result in a non-homogeneous distribution of the hydraulic conductivity and hence impacting negatively the containment safety function if they remain in the long-term. The EB experiment, even though the conditions are not completely similar to those in a real repository, has shown that the homogenization of a bentonite buffer is efficient in terms of sealing all the initial voids and providing a sufficiently low hydraulic conductivity. The uncertainties in the long-term performance of bentonite barriers have thus been reduced in some areas, related to the Spanish concept.

Case 4:

Uncertainties in Geochemical Evolution

The uncertainties identified in ENRESA (2000) and reported in D1.1 are related to the interaction process of corrosion products and bentonite: the extent of bentonite alteration due to iron corrosion, and the exact type of alteration products produced, cannot be predicted with any confidence. It was stated that current models should be tested with data from experiments and further improved. Even though the increase in knowledge obtained by the experimental work conducted in PEBS is recognized, these uncertainties are not yet completely solved. Experiments performed by CIEMAT and UAM seems to indicate that the altered zone at the canister-bentonite interface is very limited in thickness, but the impact of the iron-bentonite interaction remains uncertain, especially regarding the potential knock-on effect on the swelling pressure. Reliable long-term modelling of the process is not yet achievable because a large number of uncertainties associated with kinetics of reactions, porosity changes and thermodynamic data for Fe-silicates, still remains.

6 Summary and Future Perspectives

6.1 Summary

The PEBS project has been successful in developing an improved understanding of the early transients in bentonite barriers and reducing the associated uncertainties in the context of long-term safety assessments for disposal of spent fuel and HLW.

Within the project there were a broad range of laboratory and in situ experiments on bentonite dealing with THMC processes associated with the short-term transients in the EBS, combined with extensive modelling of these processes. New large-scale in situ studies included the HE-E experiment and the decommissioning, HM modelling and post-test analysis of the EB experiment. In addition, detailed modelling of the FEBEX mock-up, started in 1997, combined with small-scale laboratory tests, provided a good opportunity to test THM models.

In order to permit integration of a broad range of information and to put information in context, four cases were identified and the broad conclusions are briefly noted below.

Case 1:

Uncertainty in Water uptake in the Buffer below 100 °C

- There was good agreement in THM modelling between models and data for large-scale heater experiments (with high re-saturation rate/water supply), but late stage re-saturation is slower than predicted with models.
- Various model variants (double porosity, thermo-osmosis, Darcy threshold) were tested but results do not clearly permit discrimination.
- Despite this, the context from long-term safety is clearly improved - it can be stated that even though saturation is not yet fully achieved (e.g. after 15 years of FEBEX), the safety function is achieved because sufficient swelling pressure is reached throughout the barrier at 85-90 % average saturation. The model uncertainty is thus not important from a long-term safety perspective.

Case 2:

Uncertainty in the Thermal Evolution of the Buffer above 100 °C

- A new 1:2 scale URL experiment (HE-E) shows that there is reasonable agreement between models and measured TH parameters in early re-saturation; temperature field in EBS and host rock (up to 140 °C) is modelled accurately.
- Re-saturation is slow (as expected; driven by host rock water supply) and so it will require some years of monitoring to adequately test models for re-saturation.
- Further studies of the effects of heating bentonite above 100 °C in a partially saturated state suggest that the swelling pressure may be somewhat reduced (~25 %), but will still meet requirements.
- Review of process understanding and data support do not suggest important changes in performance in this high temperature range.

Case 3:**Uncertainty in HM Evolution of the Buffer**

- Cementation during heating-cooling cycle can increase strength of dense bentonite; more data has been obtained, which has shown that the effects are small below 100 °C.
- The observed cementation process is not kinetic - i.e. results are basically the same for a 1 day or long duration cycle.
- Safety relevance is related to mechanical impacts on canister (e.g. shear across a borehole); cementation also reduces swelling pressure, but has little effect on hydraulic conductivity.
- Various laboratory and field (EB) experiments show that dense bentonite pellets evolve to a swelled material indistinguishable from swelled block material from a hydro-mechanical perspective.
- The EB experiment shows that even under non-optimum emplacement conditions swelling of mixtures of blocks and pellets with large initial density differences can achieve effective sealing.

Case 4:**Uncertainty in Geochemical Evolution of the Buffer and its Interfaces with the Canister and Rock or Liner/Tunnel Support**

- The main effects of geochemical evolution are clearly at interfaces.
- Based on review of published data, below about 130 °C, limited alteration of smectite will occur within the main part of the barrier based on alteration models and natural analogues - an important factor is that the thermal phase is short.
- At a steel canister interface, the bentonite alteration is very limited over periods of several years, but is difficult to estimate over long periods.
- Impacts on system interfaces can be bounded (a few cm reaction zone over the very long term), although porosity evolution clearly requires work (both for steel-bentonite and cement-bentonite).
- It should be kept in mind that geochemical modelling provides valuable insights but is not fully predictive (especially over the long term) - a lot of model testing and supporting information is needed.

6.2 Future Perspectives

Over the next years there are good prospects for resolving residual uncertainties in the performance of bentonite barriers that relate to how the thermal and re-saturation period may affect the long-term performance. Some important experiments that will contribute to this improvement in understanding include:

- The excavation of the in situ FEBEX experiment, which will obtain valuable information on the properties of a bentonite barrier that has been exposed to partial saturation at temperatures of up to 100 °C for 15 years.
- Continuing observations in the FEBEX mock-up test over a longer period should provide relevant information on the likely mechanisms operating at the later stages of saturation.
- The HE-E experiment, which permits models for TH evolution of bentonite barriers and near-field clay rock to be compared - the slow re-saturation of the bentonite should permit

further testing of models and eventual decommissioning will permit the properties of bentonite exposed to temperatures of up to 140 °C to be assessed.

- The first two heaters in the Prototype Repository at the Äspö HRL have been excavated and the results from the investigations are about to be published. The ABM test at Äspö, where a number of different bentonites are/have been exposed to temperatures up to 130 °C for different time periods and the mineralogical effects are being investigated.
- Work on cement-bentonite and cement-host rock interfaces, in particular determination of porosity and nature of alteration is ongoing at a number of institutes and has been proposed for a European project.

In relation to lessons learned from the PEBS studies that provide relevant feedback to design, the following points are noted:

- Laboratory and field studies have shown similar performance of blocks and pellets after re-saturation, which provides some confirmation that suitable design concepts are being used.
- In terms of cement/bentonite interactions, while there are improvements in the overall chemical modelling of the associated processes, it remains difficult to constrain the long-term impacts without use of low pH cement as a design measure.
- The studies performed help to define buffer design parameters such that early re-saturation phase-induced heterogeneities in density are occurring within a range that will not violate the safety function indicators related to sufficiently high swelling pressure and low hydraulic conductivity even if it is assumed that the heterogeneities do not disappear over time.

7 References

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