### Long-Term Performance of Engineered Barriers for High-Level Waste Repositories – 15358

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#### ABSTRACT

The evolution of the engineered barrier system (EBS) of geological repositories for high-level radioactive waste (HLW) and spent fuel (SF) has been the subject of many national and international research programs during recent years. The emphasis of the research activities was on the elaboration of a detailed understanding of the complex coupled thermo-hydro-mechanical and –chemical (THM-C) processes, which are expected to evolve in the early post-closure period in the near field. From the perspective of radiological long-term safety, an in-depth understanding of these coupled processes is of great significance, because the evolution of the EBS during the early post-closure phase may have a non-negligible impact on the safety functions at later stages of the repository's lifetime. Process interactions during the resaturation phase (heat pulse, gas generation, non-uniform water uptake from the host rock) could impair the homogeneity of the safety-relevant properties in the EBS (e.g. swelling pressure, hydraulic conductivity, diffusivity).

The 7th Framework EURATOM PEBS project (long-term **P**erformance of **E**ngineered **B**arrier **S**ystems) was initiated in 2010 and completed in March 2014. The project aimed at evaluating the sealing and barrier performance of clay-based EBS over time. The project approach included experiments, model development and consideration of the potential impact on long-term safety functions.

The specific project aims were to:

- deepen the understanding of the THM and THM-C evolution of the EBS system with time,
- provide an enhanced quantitative basis for relating the evolutionary behavior to the safety functions,
- clarify further the significance of residual uncertainties for long-term performance assessment.

It can be concluded that uncertainties in process understanding occurring in the resaturation period have been better constrained through PEBS studies, thus the uncertainties in the long-term performance of bentonite barriers have been reduced in some areas. Improvements are in the areas of evolution of materials properties and model development and testing at various scales. On-going in-situ experiments in underground research laboratories may play an important role in further confirming bentonite performance over periods of 10 - 20 years.

#### INTRODUCTION

Bentonite barrier systems are proposed in repository concepts in crystalline rock, in which transport is predominantly controlled by fractures, and in clay rock, in which transport is diffusion controlled. The aim of the project PEBS (Long-term Performance of the Engineered Barrier System) is to evaluate the sealing and barrier performance of a clay-based EBS with time, through a comprehensive approach involving experiments, model development and consideration of the potential impacts on long-term safety functions, which for bentonite can be defined as containment and retardation. For a given repository concept, these safety functions can be satisfied by achieving a sufficiently high swelling pressure and low hydraulic conductivity, the quantitative requirements for which the actual values depend on the repository concept.

The experiments and models in PEBS covered the full range of conditions from initial emplacement of wastes (high heat generation and the very low EBS saturation) through to later stage establishment of near steady-state conditions, i.e. full saturation and thermal equilibrium with the host rock. The final objective of the project was to improve the treatment of the early transients in long-term safety assessments for HLW/Spent fuel. The consortium involved 15 European organizations as well as the Beijing Research Institute for Uranium Geology and covered the period 2010 - 2014.



Fig.1 Conceptual illustration of repository evolution, along with an illustration of its relationship with large-scale experiments being studied in PEBS

A conceptual illustration of repository evolution, along with an illustration of the information flow from large-scale experiments being studied in PEBS is shown in Fig. 1. The importance of uncertainties arising from potential disagreement between the process models and the laboratory and in situ experiments performed within PEBS, and their implications for extrapolation of results have been assessed, with particular emphasis on possible impacts on safety functions.

Details of the many laboratory and modeling studies and the findings associated can be found on the Project website at http://www.pebs-eu.de/PEBS/EN/Home/PEBS\_node\_en.html.

# **PROJECT METHODOLOGY**

An important aspect of developing a synthesis of modeling and experimental findings for the EBS is having a suitable framework for evaluation and integration of the diverse types of information. The PEBS project was structured to allow the national safety cases relevant to bentonite barriers to formulate the cases of interest. The cases that are defined represent a combination of a configuration (the defined EBS with its initial conditions) and the description of the evolution of the EBS reflecting the identified uncertainties of process-based transient repository evolution. The defined cases are:

- Case 1 Uncertainty in water uptake in the buffer, for the case of a maximum temperature at the canister-buffer interface of <100°C,
- Case 2 Uncertainty in temperature evolution in the buffer for the case of a maximum temperature at the canister-buffer interface of >100°C,
- Case 3 Uncertainty in HM evolution of the buffer and
- Case 4 Uncertainty in geochemical evolution of the buffer and its interfaces with the canister and rock or liner/tunnel support.

Results from the main large-scale experiments related to these defined cases, as well as from small-scale laboratory studies, are compared to coupled THMC (thermal, hydraulic, mechanical, and chemical), HM and THM models of the relevant processes in the experiments.

For each case, the importance of uncertainties arising from disagreement between models and experiments, have been assessed. In addition, the implications for extrapolation of results from various studies over the transient phase to the long-term (tens of thousands of years) for the engineered barrier system have been evaluated.

## **EXAMPLES OF STUDIES IN PEBS**

The PEBS project takes advantages of a broad spectrum of large-scale laboratory and in situ experiments, some of which have been in progress for many years.

A selection of activities is:

- the FEBEX laboratory mock-up and the associated full scale in situ experiment at the Grimsel Test Site [1], [2], [3]
- the dismantling of the full scale isothermal EB experiment at the Mont Terri URL including seismic monitoring [4], [5]
- the design and construction of the half-scale heater test HE-E at the Mont Terri URL [6] and the supporting column tests on HE-E materials [7]

- studies on stress-strain behavior at elevated temperatures [8]
- laboratory tests mimicking canister-bentonite concrete interfaces [9], [10], [11]

Extensive modeling work is performed associated with these experiments (HM modeling for the EB experiment [12], TH and THM modeling for the HE-E experiment [6] and THM-C modeling for certain laboratory experiments. An additional modeling task had the objective to use the data and improved models from the experiments for extrapolation to long-term evolution and to investigate the model uncertainty and its impact on long-term prediction [13].

Because of limited space, only two large-scale experiments can be discussed here.

#### The HE-E Heater Test at the Mont Terri URL [14], [6]

The early post-closure thermal behavior at high clay barrier temperature (see Fig. 1) is elucidated by the HE-E experiment (Fig. 2). The HE-E experiment, designed and constructed as part of PEBS, is a 1:2 scale heating experiment considering natural resaturation of the EBS at a maximum heater surface temperature of 140 °C. The experiment is performed in a 50 m long microtunnel of 1.3 m diameter located in the Opalinus Clay (OPA) of the Mont Terri URL (Switzerland). The test section of the microtunnel has a length of 10 m.

The aims of the HE-E experiment are elucidating the early non-isothermal resaturation period and its impact on the thermo-hydro-mechanical behavior, namely: (1) to provide the experimental data base required for the calibration and validation of existing THM models of the early resaturation phase and (2) to upscale thermal conductivity of the partially saturated EBS from laboratory to field scale (pure bentonite and bentonite-sand mixtures).



Fig. 2 Left: Schematic layout of the HE-E experiment in 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. Right: Picture showing the granular bentonite emplacement during the construction of the experiment.

The construction of the HE-E experiment took place between December 2010 and June 2011. In a first step a U-profile railway was installed in the 50 m long tunnel. Subsequently the host rock was instrumented. The EBS instrumentation modules were then towed into the tunnel leading to the connection of the heater liner elements into one continuous liner. An auger, adapted to the 1.3 m diameter of the tunnel, was used to emplace the granular EBS material. Emplacement densities, established during off-site tests for the MX-80 ranged around 1.45 g/cm<sup>3</sup> while for the sand/bentonite mixtures the densities were estimated to be 1.50 g/cm<sup>3</sup>. The test sections in the tunnel were separated by three concrete plugs containing also thermal insulation and a vapor barrier. Finally, in the last step, the two 4 m long heaters were emplaced in the central liner.

The instrumentation concept is targeting four zones (Fig. 2):

- 1. the heater surface where the temperature is measured;
- 2. the EBS itself and the interface with the Opalinus Clay with very dense measurements of temperature and relative humidity;
- 3. the Opalinus Clay close to the microtunnel, which was under the influence of the ventilation before and during construction where temperature, humidity, hydraulic pressure and displacement are monitored;
- 4. the Opalinus Clay at several meters from the microtunnel, where hydrostatic conditions were less disturbed by the activities in the microtunnel and where hydraulic pressures are monitored.

In the design phase it was assumed that no significant swelling pressure would develop in the EBS. A gradual temperature increase during a one year period was targeted. In June 2012 the heaters reached the maximum surface temperatures of 140°C.

During the early resaturation phase the permeability of the granular-like buffer materials is still high, but resaturation rate is limited by the low permeability of the rock, resulting in low inflow rates. Differential thermal expansion of water and solid skeleton combined with the low rock permeability is likely to induce hydraulic overpressures in the saturated Opalinus Clay at some distance from the EBS-rock interface. The measurement and assessment of these overpressures became an additional objective of the experiment.



Fig. 3: Temperature and relative humidity evolution in the middle of the bentonite section (N2) and the sand-bentonite section (G2) (Temperature (left) and relative humidity (right), in the vertical upward direction (12 o'clock) at 10 cm (C), 25 cm (M) and 45 cm (H) from the heater surface.

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 due to the low thermal conductivity of its very dry state especially in the inner part of the buffer. At the Opalinus Clay interface a temperature of below  $50^{\circ}$  C is registered. The temperature increase 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, the high sensitivity to changing two-phase flow parameters and the impact of vapor diffusion in a changing porous matrix. The vapor is driven out, in a most likely 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 vapor. The highest temperatures (above  $100^{\circ}$  C) are thus prevailing in an EBS with very low water content (below 20 % relative humidity).

The natural water inflow from the Opalinus Clay is occurring slowly through diffusion. After 32 months only at distances in the Opalinus Clay above 1 m from the tunnel wall is a positive hydraulic pressure registered. The hydraulic pressure front is progressing toward the EBS, but how long this will take and when an equilibrium state will be reached at the constant heater temperature of 140° C cannot be determined from the current dataset. This can only be assessed using modeling.

The measured temperatures and relative humidity 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 of the two at emplacement were somewhat different. This rapid homogenization can also (partly) be explained by vapor movement and has been 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

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off a couple of months after the heater temperature stabilized. These overpressures were within the expected range.

#### The EB-in situ Experiment in the Mont-Terri URL [4], [5]

The Engineered Barrier (EB) experiment represents the end of the transient phase (see Figure 1.1) when temperatures will have returned to those initially present in the host rock and full saturation has been reached.

The experiment was installed in a 6 m long section of a gallery (2.6 m high and 3.0 m wide) excavated in the Opalinus Clay of the Mont Terri URL (Fig. 4). A dummy canister (similar dimensions to the ENRESA and Nagra reference canisters) was installed on the top of a bed of bentonite blocks and the remaining open volume (approx. 28.4 m<sup>3</sup>) of the section (finally sealed with a concrete plug) was filled with a Granular Bentonite Material (GBM). An average dry density of about 1.36 t/m<sup>3</sup> of the emplaced GBM was obtained<sup>1</sup>. The experiment ran for more than ten years, under isothermal conditions and with artificial hydration during the first approx. 5 years. It was dismantled in the period October 2012 – January 2013 with more than 500 samples of the bentonite taken for on-site and laboratory analyses, which included dry density and moisture content; suction; pore size distribution; basal spacing; thermal conductivity; hydraulic and gas conductivity; swelling strain and swelling pressure, and microbial analyses.

The evolution of the GBM and the bentonite blocks were characterized geophysically. The Excavation Damaged Zone and Excavation disturbed Zone features (EDZ / EdZ) were of special interest particularly during the final dismantling process.



Fig. 4 EB experiment lay-out prior to the granular bentonite material emplacement [15]; Right : preliminary results showing the interpolated dry density (in g/cm<sup>3</sup>; in section A1-25 cross-cutting the front part of the heater dummy) measured at the samples taken during the dismantling of the EB-experiment in 2013 [4].

<sup>&</sup>lt;sup>1</sup> It should be noted that in the EB experiment there were 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.

According to the monitoring of the experiment during its more than 10 year lifetime, 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 m<sup>3</sup> (while the estimated air volume before the water injection was 12.5 m<sup>3</sup>) 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/m<sup>3</sup> a maximum swelling pressure of only 1.3 MPa was expected). All the hygrometers (except one) showed full saturation.
- After the relatively short first period (1.5 years), during the remaining lifetime of the experiment (approx. 9 years), water intake was very slow (and no additional water was artificially injected after 5 years); 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 done immediately after taking the samples, the following can be concluded:

- The average degree of saturation was 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 deformation (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 during the dismantling operation. It is thus likely that the actual degrees of saturation are even higher than those calculated from the on-site test results. It has been estimated that very probably the actual degree of saturation varies between 98 % and 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 largely diminished), the overall density range is still significant and has a variation of the order of 0.3 g/cm<sup>3</sup>. 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) as shown in Fig. 4.

The results of the laboratory tests further provided the following results related to the characterization of the dismantled bentonite barrier [16]:

- Most of the water content average values determined in the different GBM parts and blocks range between 33 % and 44 %; and the dry density varied between 1.24 and 1.42 g/cm<sup>3</sup>.
- One of the main objectives of the EB experiment was to determine if the hydraulic conductivity of the GBM (after saturation) is low enough to satisfy requirements, even if emplaced with relatively low average dry density (1.36 t/m<sup>3</sup> in this case). The permeability tests performed on fifteen samples do confirm that, after saturation, the barrier had a low hydraulic conductivity (Kw), with values that are equal to or lower than  $5 \times 10^{-12}$  m/s<sup>2</sup>.
- The measured thermal conductivity values are relatively high (as expected due to the saturation degree), ranging from 0.90 to 1.35 W/(m•K).
- Although the samples were almost fully saturated, they still exhibited some suction (remaining

<sup>&</sup>lt;sup>2</sup> There was one exception of  $8 \times 10^{-12}$  m/s, obtained with a very low dry density sample (1.18 t/m<sup>3</sup>), less representative of the overall barrier.

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).

- According to the pore size analyses, a relevant percentage of the porosity of the GBM samples was in the microporosity range (diameter smaller than 7 nm), although a macro-pore family (sizes between 3 and 35 micro meter) has also been detected, which did not exist in the original GBM pellets. Macro-porosity is predominant for samples with dry density lower than 1.30 t/m<sup>3</sup>.
- 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.

### **CONCLUSIONS AND OUTLOOK [17]**

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.

The project included 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 modeling of the these processes. New large-scale in-situ studies included the HE-E experiment and the decommissioning, HM modeling and post-test analysis of the EB experiment. In addition, detailed modeling of the FEBEX mock-up, which started in 1997, combined with small-scale laboratory tests provided a good opportunity to validate the 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. A detailed discussion of the cases can be found in [18].

Case 1 - Uncertainty in water uptake in the buffer below  $100^{\circ}$  C

- There was good agreement in THM modeling between models and data for large-scale heater experiments (with high resaturation rate / water supply), but late stage resaturation 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 interpretation in the context of 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 resaturation; the temperature field in EBS and host rock (up to 140° C) is modeled accurately.
- Resaturation is slow (as expected; driven by host rock water supply) and so it will require some years of monitoring to adequately test models for resaturation.
- 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 the 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 a 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 after a heating-cooling cycle 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 the 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 the 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 further research (both for steel-bentonite and cement-bentonite interfaces).
- It should be kept in mind that geochemical modeling provides valuable insights but is not fully predictive (especially over the long term) a lot of model testing and supporting information is needed.

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 resaturation period may affect the long-term performance. 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 resaturation, 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 modeling 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 define buffer design parameters such that early resaturation phase-induced heterogeneities in density are occurring within a range that will ensure that the safety function indicators related to sufficiently high swelling pressure and low hydraulic conductivity are in place, even if it is assumed that the heterogeneities do not disappear over time.

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