THMC behavior of clay-based barriers under high temperature – from laboratory to URL scale

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ABSTRACT

International disposal programs have been investigating if clay-based barriers can withstand temperatures higher than the 100 °C threshold for bentonite performance assumed in some advanced repository designs. For example, the United States disposal program is investigating the feasibility of direct geological disposal of large spent nuclear fuel canisters currently in dry storage.

The performance of bentonite barriers in the < 100 °C temperature range is underpinned by a broad knowledge base built on laboratory and large-scale in-situ experiments. Bentonite parameter characterization above 100°C is sparser (especially for pelletized materials), although up to about 150 °C no significant changes in safety-relevant properties are indicated[1]. At temperatures above 150 °C, it is possible that a potentially detrimental temperature-driven physico-chemical response of materials (cementation, illitization) may occur, the characteristics of which are highly dependent on, and coupled with, the complex moisture transport processes induced by strong thermal gradients. The impact of such complex processes on the performance of a repository cannot be realistically reproduced and properly (non-conservatively) assessed at the smaller laboratory scale. Such an assessment needs to be conducted by large in-situ experiments in underground research laboratories (URLs), where the most relevant features of future emplacement conditions can be adequately reproduced.

This paper gives a summary of the main findings to date on high-temperature impacts and then continues to discuss the benefits of conducting large-scale experiments at high temperature (150 °C to 200 °C) in URLs. Planning discussions for such a test at the Grimsel Test Site have been initiated.

I. INTRODUCTION

International disposal programs have been investigating if clay-based barriers can withstand temperatures higher than the 100 °C threshold for bentonite and host rock performance assumed in advanced repository designs. For example, the United States disposal program is investigating the feasibility of direct geological disposal of large spent nuclear fuel canisters currently in dry storage. These canisters typically hold as many as 32 PWR assemblies and recent designs hold even more, meaning that there is significant heat output associated with these canisters. Projections show that, by the year 2025, there will be more than 3,000 such canisters in use and that sometime before 2040 more than half of the spent nuclear fuel in the U.S. will be in dry storage[2]. Cost-effective disposal of these canisters will thus become a major part of back-end fuel management strategy. Direct disposal of these canisters, without cutting them open and re-packaging the spent nuclear fuel, is attractive for several reasons, but faces the challenge of accommodating the decay heat that will be released from the canisters. There are essentially three thermal management options (and combinations thereof): 1) longer decay storage on the surface or longer ventilation of emplacement tunnels; 2) much larger spacing between canisters and tunnels (roughly doubling the repository footprint); 3) allowing for a bentonite and host rock temperature threshold much greater than 100 °C. The third option is certainly the most cost-effective one. It is thus very important to understand whether local temperatures in the bentonite of up to 200 °C could be tolerated with no significant changes in safety-relevant properties in the engineered and natural barriers.

The performance of bentonite barriers in the < 100 °C temperature range is underpinned by a broad knowledge base built on laboratory and large-scale in-situ experiments. Bentonite parameter characterization above 100 °C is sparser (especially for pelletized materials), although up to about 150 °C no significant changes in safety-relevant properties are indicated[1]. A small number of large-scale in-situ heater tests at these higher temperatures have recently been performed or are on-going in Sweden (at the Äspö URL) and in Switzerland (at the Mont Terri Rock Laboratory). Early results from the HE-E test at Mont Terri, an experiment at higher temperatures (140 °C) representing a horizontal emplacement repository concept, suggest that important vapor movement takes place, which results in a rapid homogenization of differences in certain initial characteristics of the emplaced materials (blocks and pellets)[3]. Information on the potential impact on bentonite properties will only become available after dismantling.

Small-scale lab studies on the physico-chemical response of materials to high temperatures, such as changes in mineral chemistry or mechanical properties, have been sparse but more studies are underway. At temperatures of 150 °C and above, it is expected that significant cementation and perhaps also illitization effects will occur (although recent results indicate that high potassium concentrations are required), which could affect the swelling and perhaps also the sorption capability of the bentonite. In addition to the physico-chemical response of materials to high temperatures, strong thermal gradients will also induce complex moisture transport processes, including convection of vapor, the combined impact of which can only be assessed at the large (underground research laboratory) scale since these regimes cannot be reproduced at the much smaller laboratory scale.

II. SAFETY FUNCTIONS AND REQUIREMENTS OF CLAY-BASED ENGINEERED BARRIERS

The criterion for evaluating the overall performance and safety of a repository is formulated as a maximum dose or risk to a representative individual in the group exposed to the greatest risk. It is issued by the national regulators and is evaluated on the basis of the Safety Case, which includes numerical safety analysis calculations. The concept of the safety function has been introduced to allow the individual repository components and how they contribute to overall repository safety to be studied in a more practical way, together with the safety requirements for containment, isolation, retardation, attenuated release and low release rates.

For the buffer materials in the case of HLW/SF disposal, the key requirements are low hydraulic conductivity, self-sealing ability and durability of the properties on the very long-term[4]. These have led to the selection of clay or clay-based materials for the buffer, with the following safety functions[4]:

- Limiting transport in the near-field: this is primarily limiting advective transport (determined by the hydraulic properties), but also retardation or attenuation (determined by the chemical retention properties)
- Reducing microbial activity (in order to protect the canister)
- Damping rock-shear movements (in order to protect the canister)
- Resisting mineralogical transformations (in order to retain the favorable properties on the short- and long-term)
- Preventing canister sinking (in order to protect the canister)
- Limiting pressure on canister and rock (in order to protect the canister or the overlying bedding structure in the case of sedimentary host rocks)

For sedimentary host rocks, an additional safety function of the buffer, in the context of attenuated release discussed above, is for the buffer to exert sufficient pressure on the host rock to prevent an extension of the EDZ and enhance its self-sealing capacity.

Two physico-mechanical properties of clay-based engineered barriers, which can be used as indicators for the fulfillment of most of the safety functions listed above, are the swelling capacity and the hydraulic conductivity. These depend on the type of clay used for the buffer, as well as the density of the buffer at emplacement. High temperature effects, such as potential cementation or illitisation of bentonite, possibly affecting the target values required to fulfill the safety functions (safety function indicator criteria), can be evaluated by assessing their impact on the swelling capacity and the hydraulic conductivity.

III. CLAY-BASED BUFFER RESPONSE ABOVE 100 °C - LABORATORY SCALE TESTS

Two different phases are recognized in the evolution of the buffer in repositories in the saturated zone, namely an initial, relatively short one during which the buffer is under partially saturated conditions and a second, longer one, during which the buffer has reached full saturation. The second phase corresponds to the long-term behavior considered in the safety analysis. The brief summary of the laboratory tests below is divided in these two phases.

III.A Thermo-hydro-mechanical-chemical properties under fully saturated conditions

A series of laboratory tests for temperatures above 100°C has been performed in the recently completed EU 7th Framework project PEBS[1]. Two different types of bentonite have been studied, MX-80 (Na-dominated Wyoming bentonite produced by American Colloid Co.) and 'FEBEX' (Mg-Ca-dominated Almeria bentonite).

Villar and Gómez-Espina (Ref. 5) and Villar et al. (Ref. 6) observed that, with respect to the FEBEX bentonite, there was a slight decrease in the swelling capacity for temperatures close to 100 °C and a decrease in the order of 30% (a very broad indicative value based on extrapolated trends) in the swelling pressure. The water-saturated permeability of the FEBEX bentonite increased with temperature, more or less as expected from the change in water viscosity. In addition, the water retention capacity of the bentonite (both FEBEX and MX-80) decreased clearly with temperature, starting at temperatures of above 60 °C, and when the density of the bentonite was high and the water content low[7].

When analyzing samples from large-scale field tests subjected to higher temperatures, some deviations in test results with respect to stresses and strains at failure or hydraulic conductivity were observed in shear tests compared to reference tests[8, 9, 10]. It could not be excluded that the deviations were caused by the increased temperature.

A study of stress-strain properties was performed in a test series in which specimens of MX-80 were exposed to 90 °C, 120 °C and 150 °C and specimens of FEBEX bentonite were exposed to 90 °C (Ref. 11). The heating for 24 hours in a laboratory oven was done both before and after full saturation. Heating after full saturation was at a controlled water pressure on the top of the specimen (600 kPa) and that before full saturation under sealed conditions but without pressure control. The influence of increased temperature was quantified by measurements of stresses and strains during unconfined compression tests and also by measurements of swelling pressure and hydraulic conductivity made after heating. The results showed i) a tendency for increased deviator stress at failure with increased temperature, with significant deviations after heating to 120 °C and 150 °C, ii) a significant decrease in strain at failure after only short-term heating to 150 °C and iii) small decreases in swelling pressure and hydraulic conductivity.

Laboratory experiments under fully saturated conditions have also been conducted with the focus on the degree of temperature-driven illitization in bentonite. These experiments typically involved hydrating bentonite samples with various types of solutions and exposing them to different temperature conditions for a given time period. The solutions used in the experiments differed in the concentration of Na and K, and the test temperatures ranged from 90 °C (Ref. 12) to 300 ºC (Ref. 13). No transformation from smectite to illite has been reported[12, 13, 14, 15], with one exception: A test on Wyoming bentonite using high K solution and heating at 300 ºC for 6 weeks[15] showed partial dissolution of smectite and formation of minor amounts of illite due to crystallization. It is noteworthy that Mosser-Ruck and Cathelineau[14] observed transformation of low-charge smectite to high-charge smectite, which can be a precursor to the formation of illite. Other chemical changes have been observed, including dissolution of calcite, feldspar[13], pyrite and clinoptilolite[15] and precipitation of quartz, zeolite and analcime[15]. An experiment reported in Allen et al.[16] cited in Ref. 17 is probably the longest-duration test — bentonite reacted with basaltic groundwater for a time period of 379 days at 300 ºC; smectite was not structurally changed, but compositionally altered.

III.B 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/m^3 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 undersaturation at emplacement and the heat generated by the disposal canister, drying of bentonite will occur close to the canister. Higher temperature means more drying and stronger vapor transport away from the heat source. At the same time, the inflow of water from the rock will gradually increase the moisture content of the bentonite and a saturation front will gradually move towards the canister. The timescale at which full saturation will be achieved depends very much on the temperature evolution near the canister and the magnitude of flow at the rock/bentonite interface. Results also show that the radial homogeneity of the moisture content in the buffer during resaturation is related to the degree of localized inflow. For example, in the FEBEX in situ experiment, where no strong localized flow occurs, concentric rings of relatively uniform moisture content are observed after 15 years, ranging from 100 % saturation near the rock to 80 % saturation near the heater[18]. In contrast, strongly localized fracture inflow can 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 distribution similar to the FEBEX case[3].

The consequences of the slow inflow and high 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 summarized from Johnson et al. (Ref. 1) hereafter.

Several studies on the effect of unsaturated conditions on bentonite swelling capacity were performed after the publication by Couture (Ref. 17) of results showing that MX-80 bentonite powder experiences a large reduction in expandability after treatment with steam at 150-250 °C. Oscarson and Dixon (Ref. 19) also found similar results for uncompacted Avonlea bentonite (80 % montmorillonite) down to temperatures as low as 110 °C and noted that the material would re-swell when ultrasonically treated and metaphosphate was added. Results of a more comprehensive series of experiments of the type performed by Couture are discussed in Ref. 20. Large differences in expanded volume between reference and vapor-exposed material were found in some bentonites when the bentonites were actively dispersed prior to testing. The results were qualitatively similar to what had previously been reported by Couture. However, the findings concern only the ability to form much expanded gels and the short- term exposure to water vapor at temperatures of 150 and 200 °C did not significantly change the swelling capacity.

Oscarson and Dixon (Ref. 21) also performed studies on bentonite compacted to dry densities of 1,000 to 1,300 kg/m^3. Samples were compacted with water to give initial saturation levels of 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 and the results have thus been attributed to aggregation and/or cementation phenomena.

Further work was done by Haas et al. (Ref. 22) and Madsen (Ref. 23), who passed steam through compacted samples with dry densities of 1,400 and 1,750 kg/m^3 at 150 °C. For a density of 1,400 kg/m^3, swelling pressure decreased 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 conditions in a repository.

Pelletized granular bentonite was studied by Pusch et al. (Ref. 24), who found that dense bentonite pellet samples experienced little reduction in 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 vapor. Valter and Plötze (Ref. 25) observed in a series of experiments that some reduction in swelling occurred under saturated conditions, largely in the case of samples heated to above 120 °C.

All the studies discussed above note that there is no detectable mineralogical change in thermally treated material up to 150 °C; instead the property changes seem to be related to physical aggregation and possibly some silica cementation. Material is observed to re-swell only when sonicated, usually also in the presence of phosphate. The studies include short-term experiments (as little as a few days) and a longer-term experiment (more than six months). No appreciable differences in property changes appear to exist in relation to the different durations of treatment.

Because laboratory tests aimed at chemical alteration of bentonite at high temperature were mostly conducted under saturated conditions, experimental data regarding mineralogical transformations in unsaturated bentonite are relatively sparse. In general, smectite-to-illite alteration rates can be considerably slowed depending on the saturation state. This was shown by the experimental study of Whitney (Ref. 26) indicating retardation of illitization under unsaturated conditions, a result that was attributed to the restricted transport of solutes. Recently, Caporuscio et al. (Ref. 27) heated a bentonite/water mixture at 300 °C for 6 weeks with a water to bentonite ratio of 1:5 and results showed partial dissolution of smectite.

III.C Discussion

The laboratory test results discussed above, although not completely consistent, show in general that significant reduction in swelling pressure can occur in some cases above 110 - 120 °C, to values of more than 50 % of the initial values. The reasons for inconsistency of the 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 resaturation 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.

Whereas smectite-to-illite transformation has been widely observed in geological systems[28], this mineralogical alteration has rarely been seen in laboratory experiments. Cheshire et al. (Ref. 15) is probably the only test that shows partial dissolution of smectite and crystallization of illite. A couple of factors might explain this discrepancy. The first is related to reaction kinetics. Based on a widely used illitization kinetic model, it takes about 100─200 years for about 1% illitization to occur, which means most laboratory studies are simply not long enough to generate detectable mineral changes. Second, the formation of illite is only one chain of a complex reaction network. Some test conditions are not favorable for illitization, including the lack of Al and K and excessive Na, Ca, Mg and Si.

IV. CLAY-BASED BUFFER RESPONSE ABOVE 100 °C - MODELING STUDIES

Only a few modeling studies simulated chemical and mechanical alterations at temperatures significantly above 100 °C; among them are those described Liu et al. (Ref. 29) and Zheng et al. (Ref. 30). These models focused on the chemical alteration in EBS bentonite and the subsequent stress changes caused by high temperature and chemical changes. In this section, we briefly summarize their modeling scenario and findings.

IV.A Summary of recent modeling results

In the models presented in Ref. 29 and 30, coupled THMC processes associated with a hypothetical bentonite-backfilled nuclear waste repository in clay rock were simulated using TOUGHREACT-FLAC, a simulator that sequentially couples the multiphase fluid flow and reactive transport simulator TOUGHREACT[31] with the finite-difference geomechanical code FLAC3D[32]. The repository example involves a horizontal waste emplacement tunnel at 500 m depth[33]. In these generic cases, it is assumed that the EBS backfill is composed of either Kunigel-V1[34] or FEBEX[35] bentonite, and that the host rock properties are representative of Opalinus Clay[36, 37]. The illitization is simulated via smectite dissolution and illite precipitation. The kinetic law for mineral dissolution/precipitation is given in Ref. 31, which follows the transition state theory (TST) or TST-like equations[38]. The illitization rate (the rate of illite precipitation and smectite dissolution) was calibrated based on the measured illite percentage in an illite/smectite (I/S) mixed layer from Kinnekulle bentonite, Sweden[28]. An extended linear elastic swelling model was used to take into account the effect of changes in the abundance of swelling clay (smectite) and ion concentration in pore water on the swelling stress[29].

Two scenarios were simulated for comparison: a case in which the temperature in the bentonite near the waste canister can reach about 200 °C and a case in which the temperature in the bentonite near the waste canister peaks at about 100 °C. In addition to the base case scenarios, sensitivity analyses for the key chemical and mechanical parameters were conducted to evaluate the key controlling factors of illitization such as the aqueous composition of pore water in EBS bentonite and host rock and the dissolution/precipitation of accessory minerals such K-feldspar and quartz.

Modeling results indicate the occurrence of some degree of illitization in the EBS bentonite. Other chemical alterations include the dissolution of K-feldspar and calcite, and the precipitation of quartz, chlorite and kaolinite.

In general, illitization in the bentonite is enhanced at higher temperature. However, the extent of illitization, expressed as the smectite volume fraction change, is affected by many chemical factors and consequently varies a great deal, which could be the reason that illitization is not consistently observed in laboratory studies. The concentration of K is unsurprisingly the most important chemical factor. However, the dissolution of K-feldspar within the EBS rather than the transport of K from the host rock to the EBSis the major source of K for the illitization in EBS bentonite. As a result, the amount of K-feldspar in the EBS bentonite and its dissolution rate strongly affect the degree of illitization. Models also show that a high concentration of Na in the bentonite could suppress the illitization, which suggests that the use of a bentonite mixture with a higher Na concentration could be a mitigating measure if illitization is a concern for higher temperature repository designs.

The models [29, 30] also show that higher temperature may lead to higher stress in the near-field, caused by thermal pressurization of pore water in the low-permeability rock surrounding and connected to the emplacement tunnel. During this period of strong thermal stresses, smectite dissolution causes only minimal changes in the simulated total stress. Chemical changes, including changes in pore water ion concentration and smectite volume fraction, lead to a reduction in swelling stress.

Overall, modeling studies suggest that illitization is a complex process impacted by several hydrogeological and chemical characteristics. Several factors could reduce or prevent the occurrence of illitization, for example: i) limited supply of K; ii) high volume fraction of Ca, Mg and especially Na in a water-bentonite system; iii) production of excess silicate and precipitation as quartz; iv) availability of Al; and v) dry conditions caused by heating (which would be the condition existing at the waste package surface during early repository stages).

IV.B Discussion

Simulating the coupled THMC alteration in EBS bentonite is very challenging and several important uncertainties remain. First, the illitization rate at very high temperatures needs to be better constrained. In geological formations, studies[39] showed that the onset of naturally occurring smectite illitization can occur in a temperature range from ~70 °C to ~100 °C, which is also manifested by the fact that the smectite fraction in an illite/smectite mixture decreases sharply at a depth of 2500 m to 3000 m (Ref. 40). Data on naturally occurring illitization above 100 °C are fairly sparse, although a few studies are available in the literature[41]. Laboratory experiments at high temperature can be helpful in constraining the illitization rate, but caution has to be exercised because it is known that the reaction rate measured in the laboratory could be orders of magnitude higher than that observed in the field[42]. Large-scale (and preferably long-term) experiments at very high temperatures will provide the most reliable data on illitization at high temperature.

Second, the effect of moisture movement on illitization needs to be better understood. While most studies target the chemical aspects of illitization, hydrodynamic constraints deserve more studies, as illitization at high temperature can be significantly limited, especially in the area near the heater. Higher temperature leads to stronger moisture movement and therefore more pronounced evaporation. This inhibits the dissolution of K-feldspar, which in turn limits the supply of K and suppresses illitization. Evaporation can also cause the precipitation of some minerals (e.g. calcite) and lower the porosity and therefore limit the transport of key ions such K and Al from the surrounding host rock.

Third, the mechanical effects of high temperature on EBS bentonite need more investigation. Models from Ref. 29 and Ref. 30 show that thermal pressurization in the low-permeability rock surrounding and connected to the emplacement tunnel can cause much higher stress in the near-field.

V. CLAY-BASED BUFFER RESPONSE ABOVE 100 °C - LARGE SCALE FIELD TESTS

Only a few large-scale *in situ* tests have been conducted specifically targeting higher temperatures at or above 100 °C. Among the reasons for this are that, from early on, most repository concepts were designed with limiting the canister surface temperature below 100 °C and that setting up such large-scale tests, particularly long-term ones, requires a considerable commitment of resources. In the following, two of the on-going large-scale heater experiments are highlighted.

V.A HE-E experiment

The HE-E experiment[3] represents a first large-scale test of the understanding of thermo-hydraulic behavior of the Nagra near-field concept. It is a 1:2 scale heating experiment considering natural resaturation of the EBS and a maximum heater surface temperature of 140 °C. The experiment is located in the Opalinus Clay of the Mont Terri URL (Switzerland) in a 50 m long microtunnel of 1.3 m diameter (Figure 1). It was initiated as part of the PEBS project (long-term performance of the engineered barrier system), a 4 year project within the EU 7th Framework Program.

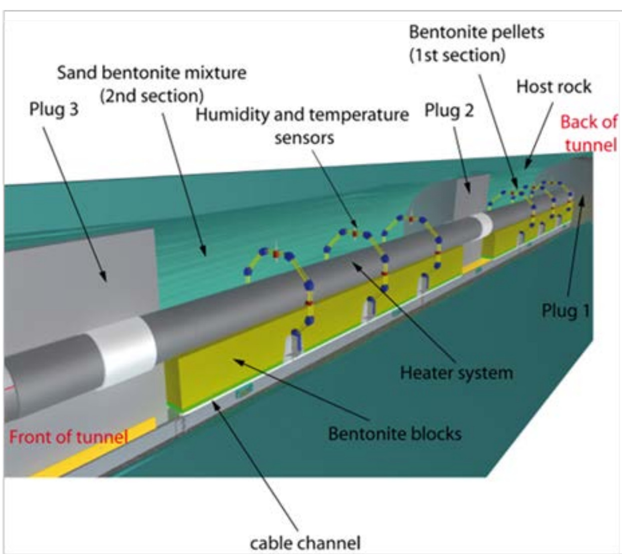

Figure 1. The HE-E experiment layout

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

The experiment consists of two independently heated sections of 4 meters each, whereby the heaters are placed in a steel liner supported by MX-80 bentonite blocks (dry density 1,800 kg/m^3, water content 11%). The two sections are fully symmetric apart from the granular filling material. While section one is filled with a 65/35 granular sand/bentonite mixture, section two is filled with pure MX-80 bentonite pellets. The construction of the HE-E experiment took place between December 2010 and June 2011. An auger, adapted to the 1.3 m diameter of the tunnel, was developed to emplace the granular EBS material. Emplacement densities, established during off-site tests for the MX-80, were around 1,450 kg/m^3, while for the sand/bentonite mixtures these were estimated to be 1,500 kg/m^3. Observations and results from the first 2.5 years of the experiment are summarized below.

The temperature increases observed in the EBS and Opalinus Clay were in line with those predicted by the design calculations. 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-EBS interface, a temperature below 50 °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 in 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 from the heat source, 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 vapor. The highest temperatures (above 100 °C) prevail in an EBS with very low water content (below 20% relative humidity).

The natural water inflow from the Opalinus Clay occurs slowly through diffusion. After 32 months, only at distances in the Opalinus Clay more than 1 m from the tunnel wall is a positive hydraulic pressure registered. The hydraulic pressure front is progressing towards 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 both 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 experiment, as this is the first large-scale, high temperature experiment with different materials.

V.B FEBEX experiment

The FEBEX experiment[18] in the Grimsel Test Site (Switzerland) consists of a full-scale in situ engineered barrier system (EBS) test for the disposal of high-level waste (HLW). It is being performed under natural conditions in crystalline rock in which the dummy canisters are placed horizontally in drifts and are surrounded by a clay barrier constructed of highly compacted bentonite blocks (Figure 2). Heating started in 1997 and, since then, a constant temperature of 100 °C has been maintained in the remaining heater, while the bentonite buffer has been slowly hydrating in a natural way. Partial dismantling and sampling of one section of the experiment was carried out during 2002.

The hydration pattern is relatively symmetric, with no major differences along the axis. Although the host rock is characterized by heterogeneities with zones of higher permeability, the resaturation process is driven by the suction of the bentonite rather than by the availability of water in the rock, especially in the early phase. After 17 years, the water content in the buffer close to the heater still continues to increase slowly. The hydraulic pore pressures in the buffer and the geosphere have practically stabilized. The total pressure in general continues to increase in most points into the buffer, where pressures of over 6 MPa are registered in some parts.

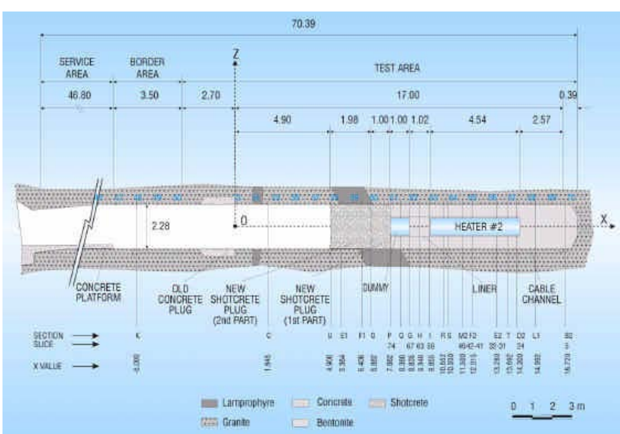

Figure 2. The FEBEX in situ test layout for the second operational phase

After 18 years of operation, the experiment will be excavated in 2015 (Project FEBEX-DP, Figure 2) and the focus of the sampling will be on[43]:

- Determination of the distribution of key physical properties, such as density and water content
- Characterization of corrosion and microbiological processes on the heater, instruments and coupons resulting from evolving redox conditions and saturation states, including gas analysis
- Characterization of mineralogical interactions at material interfaces (e.g. cement-bentonite or iron-bentonite, rock-bentonite) by macro- and micro-level studies

In addition, the excavation will allow an assessment of the sensor performance and the results will contribute to further increasing the understanding of the thermo-hydro-mechanical (THM) and thermo-hydro-chemical (THC) processes through integration of monitoring and in-situ measurements.

VI. CLAY-BASED BUFFER RESPONSE ABOVE 100 °C – FUTURE R&D STEPS

More research is needed to fully understand long-term exposure of clay-based EBS systems to temperatures of up to 200 °C to allow reliable repository performance predictions to be conducted for such conditions. Results from laboratory studies have demonstrated that strong heating can result in reduction of swelling pressure, whereas mineral transformations (smectite-to-illite transformation) have not been consistently observed. However, most laboratory experiments have been conducted over short time periods (compared to the slow reaction rate) and have not explored the full complexity of transient boundary conditions in temperature, saturation, stress, water chemistry experienced in an actual repository setting. In situ field experiments, on the other hand, have not tested the full temperature regime of up to 200 °C and have not focused on the possibility of mineralogical alterations. This may be the reason why illitization has not been conclusively confirmed for EBS bentonite in either small-scale laboratory experiments or large-scale mockup and field tests, whereas this process has been widely observed in geological systems[28] (that may or may not be representative of repository conditions) and was also shown to occur (under certain conditions) in recent modeling work. Seemingly contradictory conclusions have been reached by geological data and models versus existing laboratory and field tests. We propose here that a 1:1 scale multi-year experiment conducted at a higher (~200 °C) temperature range should be the centerpiece of a future R&D program, when expecting high nearfield temperatures, complemented by targeted laboratory tests and integrated modeling and validation work. In addition to illitization, a designated large-scale high-temperature event can also identify important THMC coupling phenomena on a realistic scale and under representative repository conditions.

The example of FEBEX discussed above demonstrates the feasibility of conducting large-scale long-term THMC experiments under repository-relevant conditions. A similar experiment, based on the Nagra concept (1:1 scale, multiple heaters, test of EBS under thermal loading) is under construction at the Mont Terri laboratory (full-scale emplacement test – FE[44]). The heating phase was initiated at the end of 2014, aiming to reach temperatures at the surface of the heaters of 140 °C.

Potential options for a targeted high-temperature experiment (150 °C to 200 °C) in a fractured rock environment are currently being considered. One possibility would be to use the well characterized FEBEX drift at the Grimsel Test Site and to bring in the knowledge of EBS behavior from the large-scale experiments discussed above[43]. Design characteristics for such an experiment are open for discussion, for example type of bentonite, target temperature, duration and additional processes to be investigated (gas migration).

The benefit of such a large-scale test, accompanied by a systematic laboratory program and modeling effort, is that the temperature effects can be evaluated under realistic conditions of strong thermal, hydraulic and density gradients, which cannot be reproduced in the laboratory. This spatial upscaling and model verification (or further development) will enhance the ability to also upscale in time and assess the high temperature effects on the EBS for relevant periods. This will lead to improved mechanistic models for the prediction of temperature-induced processes, including chemical alteration and mechanical changes, which can then be used for performance assessment (PA) analysis of high-temperature scenarios. The key question is whether higher repository temperatures would trigger mechanisms that compromise the various barrier functions assigned to the engineered components (specifically the bentonite buffer) and host rock. If the barrier function is (partially) compromised, PA analysis can evaluate whether reduced performance of a sub-barrier (or parts thereof) would still give adequate performance.

VII. CONCLUDING REMARKS

This paper gives a summary of the main findings to date on the behavior of clay-based barriers at high temperatures (higher than 150 °C) and then continues to discuss the benefits of conducting large-scale high-temperature experiments in URLs. Planning discussions for such a test at the Grimsel Test Site have been initiated. For concepts under development, a 1:1 scale experiment at higher temperatures would provide a valuable dataset, complementing a detailed laboratory program targeting a significantly higher temperature range than that currently considered as a typical threshold for many repository designs (~100 °C). For existing advanced nuclear waste programs, it would increase confidence in the available laboratory datasets if it can be demonstrated at the 1:1 scale (under conditions similar those experienced in a geological repository) that THMC effects at elevated temperatures of 150 °C to 200 °C do not significantly affect the bentonite properties.

ACKNOWLEDGMENTS

Partial funding for this work was provided by the Used Fuel Disposition Campaign, Office of Nuclear Energy, of the U.S. Department of Energy under Contract Number DE-AC02-05CH11231 with Lawrence Berkeley National Laboratory.

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