Evaluation of temperature-induced effects on safety-relevant properties of clay host rocks with regard to HLW/SF disposal

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ABSTRACT

DBE TECHNOLOGY, BGR and GRS are developing a methodology to demonstrate the safety of a repository for high-level waste and spent fuel (HLW/SF) in clays according to the requirements of the German regulating body. In particular, these requirements prescribe that the barrier effect of host rocks must not be compromised by a thermal impact resulting from HLW/SF emplacement. To substantiate and quantify this requirement, we carried out a literature survey of research on thermally-induced changes on clay properties. Effects thus compiled can be divided into thermo-hydro-mechanical and chemical-biological-mineralogical effects and were analysed with regard to their relevance to the integrity of clay host rocks. This analysis identified one effect of major influence within each group: thermal expansion and compaction as well as results of microbial activities. Importantly, it further revealed that a moderate temperature increase above 100°C cannot be expected to compromise the integrity of the geological barrier according to the current knowledge state. Evidence is presented in this paper that temperature increases up to 150°C can actually contribute to an improved performance of a radioactive waste repository by increasing the consolidation of the clay and sterilizing the repository's near-field to depress the deteriorative microbial effects. A quantitative temperature criterion for thermal impact of HLW/SF on clay host rocks is accordingly proposed.

Introduction

An integrated methodology to demonstrate the safety of a HLW/SF-repository in clays in Germany is being developed by DBE TECHNOLOGY, BGR (Bundesanstalt für Geowissenschaften und Rohstoffe) and GRS. The strength of this new integrated methodology lies in the link between legal and geological boundary conditions, disposal and closure concept, demonstration of integrity of the geological and geotechnical barriers, and long-term analysis of probable and less probable repository evolutions.

One key element is the demonstration of the integrity of the geological barrier. The regulating body requires a demonstration of the integrity of the containment-providing rock zone within host rocks for a period of 1 million years by meeting different integrity criteria (BMU, 2010). In particular, one of these criteria prescribes that the barrier effect of host rocks must not be compromised by temperature effects resulting from HLW/SF emplacement.

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In order to be able to carry out such a demonstration which necessarily involves a model effort, this qualitative requirement needs a quantification to identify the corresponding limiting temperature (temperature criterion).

Effects of temperature on host-rock properties

A key question in this relation concerns a definition of the compromising thermal impact on clays. To answer this question, a literature survey was carried out focusing on research of thermally induced changes of clay properties. The effects thus compiled have been categorized into thermo-hydro-mechanical and chemical-biological-mineralogical effects and were analysed with regard to their relevance to the geological barrier integrity as described below. A more detailed description prepared by the authors in the course of the study will be published as a technical report following this publication (Jobmann *et al.*, 2015).

Thermo-hydro-mechanical effects

Laboratory tests with saturated Boom Clay (Sultan *et al.*, 2002; Baldi *et al.*, 1991) showed in accordance with earlier observations for other saturated clays (Tang *et al.*, 2008) that heating of compacted or natural clay causes the clay to contract if the overconsolidation ratio (OCR, a ratio of vertical effective pre-consolidation stress to the current effective stress) is low (≤ 1). At high-OCR values, an initial period of thermal expansion turns into contraction upon exceeding a specific temperature as shown in Fig. 1.

While thermal expansion of clays takes place due to a thermal expansion of the clay matrix and of pore water, thermal contraction of clays is due to mechanical reduction of the intergranular friction. As a result of this compaction, the strength of the clay increases irreversibly (Tang *et al.*, 2008). The mechanical reduction of the intergranular friction can indeed involve the release of water adsorbed between layers and on the surface of clay minerals. The initial expansion of the material at increasing temperatures due to the expansion of the mineral matrix and of the pore water is followed by a phase where expansion and contraction counterbalance each other until the release of water and thus the contraction (consolidation) prevails (Jobmann and Polster, 2007). But even if the claystone is nearly saturated or almost undrained, contraction takes place if the temperature at the expansion-contraction-threshold (ECT) is exceeded (Zhang *et al.*, 2010).
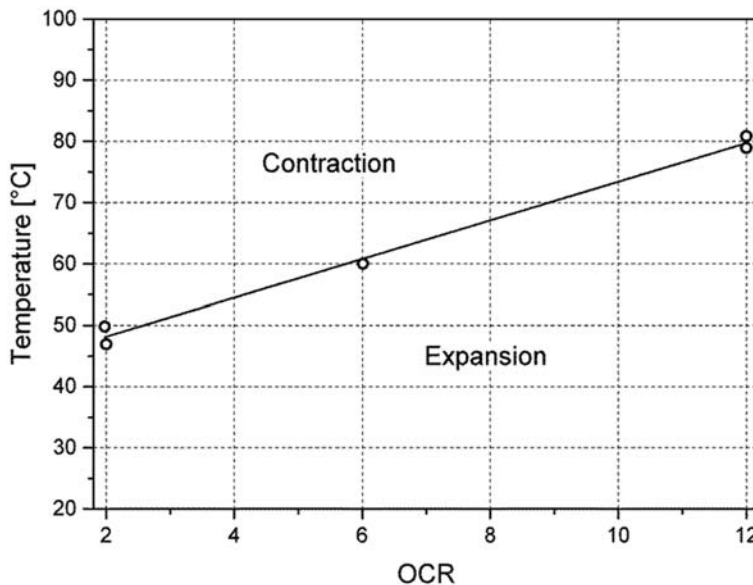

FIG. 1. Temperature at the expansion-contraction-threshold as a function of overconsolidation ratio (OCR) for Boom Clay (modified after Sultan *et al.*, 2002; Baldi *et al.*, 1991).

The ECT also depends on the stress state and suction. Following an increase in intergranular friction with high-suction tension the ECT shifts to higher temperatures. This is consistent with the results from triaxial tests using core samples of Callovo-Oxfordian clay (Zhang *et al.*, 2010) in which thermal expansion took place at 90°C in drained samples (higher suction tension) and thermal contraction took place in undrained samples (lower suction tension). The increasing effective stress, on the contrary, reduces the ECT. Experiments of Faulkner and Rutter (2003) with clay samples from fault gouges for example, document an ECT decrease from 80°C at 25 MPa to 40°C at 50 MPa. Importantly, the thermal contraction in clay persists even after subsequent cooling, while thermal expansion is reversible (Tang *et al.*, 2008).

During the large-scale *in situ* test in Boom clay, a shift from expansion at OCR = 6 to compaction at OCR = 2 to an even stronger compaction at OCR = 1 were observed between 20–95°C (François *et al.*, 2009). As a result of this compaction, the strength of the clay and the pre-consolidation pressure in the clay increased irreversibly. From heater test results in Opalinus clay, it was concluded that neither the heating nor the subsequent cooling caused thermally induced cracks in the clay (Kull *et al.*, 2007; Zhang *et al.*, 2007).

Mechanical properties of Opalinus clay obtained at temperatures between 60 and 80°C revealed a slightly reduced breaking strength, residual strength and Young's modulus when compared to results obtained at 20 to 35°C (Gräsle and Plischke, 2010). However, these laboratory tests were carried out in a temperature range that is below the ECT, i.e. in the expansion phase. A thermal contraction, in contrast, generally increases the strength and rigidity of clays. Zhang *et al.* (2010) analysed rock samples of the Callovo-Oxfordian clay at temperatures between 90 and 150°C, which is above the ECT, and found that the strength of the clay had increased significantly under thermal load.

With regard to hydraulic properties, laboratory experiments on clay samples taken from clay-bearing fault gouges showed that water permeability increases with increasing temperature from 20°C to 80°C. A further increase of the temperature above 80°C up to 150°C resulted in a continuous decrease of the water permeability (Faulkner and Rutter, 2003). The behaviour in the lower temperature range <80°C was assumed to be caused by the increasing de-stabilization and desorption of water adsorbed at the mineral surfaces which reduced the water permeability. At higher temperatures, the compaction process is dominant, leading to a reduction in porosity and permeability. At effective stresses below 75 MPa, no changes in water permeability were observed in the low-temperature range <80°C which was assumed to be due to pore dilation. Even at such relatively low effective stresses, a continuous decrease of the water permeability was observed at temperatures higher than 80°C. These observations are in agreement with laboratory experiments on Callovo-Oxfordian clay (Zhang *et al.*, 2010).

Heating of saturated clay leads to an increase in pore pressure due to the thermal expansion of the pore fluid. Due to the increase in pore pressure in clay host rocks, the resistance to gas penetration is increased as well, because the gas has to overcome this increased resistance before it can penetrate the rock mass.

Additionally, an increase in pore pressure generates hydraulic pressure gradients, causing the pore fluids to migrate. The rate of the migration process and thus of pressure decrease depends on the permeability of the clay (Jobmann *et al.*, 2007; Zhang *et al.*, 2010). This process is temporary and reversible as long as no fractures are induced. Experience from the *in situ* heater experiments shows that pore pressure decreases over a period of approximately one year without inducing any fractures.

Water in the host rock next to the waste vapourizes after exceeding the boiling point. Taking into account the hydrostatic pressure at the emplacement level, it becomes clear, however, that the boiling point under emplacement conditions of an overburden pressure of ~7 MPa will be at ~280°C. Even a small fluid pressure increase of 0.5 MPa after sealing, for instance, increases the boiling point to ~150°C.

As for the thermal properties of clay, the contraction in Opalinus clay results, for example, in a slight increase in thermal conductivity at temperatures above 110°C, which is due to a closer contact in the mineral matrix and the reduced pore space (Jobmann and Polster, 2007).

Chemical, biological and mineralogical effects

A temperature-dependent conversion of smectite to illite can considerably deteriorate the retention properties of clay. It is important to note that the smectite-to-illite reaction requires potassium to proceed. Huang *et al.* (1993) showed that for a K content of 200 ppm, which is characteristic of clays, an illitization of 80% of smectite cannot be

accomplished before 100,000 years at ~150°C (Fig. 2). The latter study concluded that the observations by Pytte and Reynolds (1989) of shorter smectite-to-illite reaction times for contact metamorphic fluids (~3500 years at ~150°C for an illitization of 50% of smectite) must be due to an order of magnitude higher K content of >2000 ppm, which is characteristic of hydrothermal fluids. Considering that the thermal impact of the emplaced HLW/SF becomes negligible after about 10,000 years (Jobmann *et al.*, 2015), being constrained to several metres around individual boreholes or tunnels with HLW/SF even at the temperature maximum, no significant illitization within clay host rocks can be expected to occur.

Dissolution of SiO_2 in clay represents a further possible effect of heating, which at subsequent cooling is followed by its re-precipitation, leading to a cementation of pores and influencing plasticity, hydraulic conductivity and swelling capacity of clay (Wersin *et al.*, 2007). The latter study concluded however that this effect causes no or only minor changes of these clay properties at temperatures of <130°C.

Thermo-chemical sulfate reduction is proved to occur during geological times at temperatures >80°C. At 90°C, the extrapolated half-life of the sulfate reduction is ~210,000 years (Truche *et al.*, 2009).

According to Cross *et al.* (2004) half of sulfate in clay pore water can be reduced to corrosive hydrogen sulfide within about 1650 years at 150°C or within ~20,000 years at 125°C. Assuming that the temperature of the host rocks will not exceed 150°C at any place, the time period of temperatures above 80°C in the host rocks will be a few hundred years only. Thus, no significant sulfate reduction is to be expected and will be limited to a relatively short time period and to the closest vicinity of emplacement drifts or boreholes.

Quantitative analysis of retention behaviour of clay at increased temperatures can be derived from experimental values of sorption capacities or distribution coefficients (Kd) of radionuclides for individual minerals constituting the clay. Very limited available data on distribution coefficients for clay minerals and dependence on temperature reveal that Kd increases for Ni, Eu, and other lanthanides and slightly decreases for Cs for temperature increasing from 20 to 150°C (Jobmann *et al.*, 2015). Since Cs is not a dose-relevant nuclide for a deep geological disposal in clays and the temperature increase is limited to a few thousand years this effect can be judged to be irrelevant.

Transformation of organics to raw oil and natural gas was recently studied using a numerical model calibrated by measured vitrinite reflectance, which
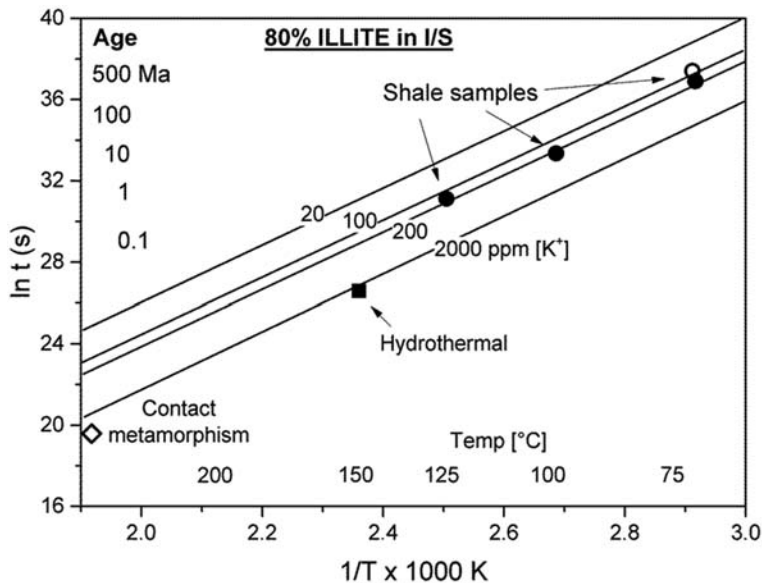

FIG. 2. Evolution of the smectite content in mixed-layer illite/smectite with time depending on the clay temperature and potassium content (From Huang *et al.* (1993, figure 7). Reproduced with kind permission of The Clay Minerals Society, publisher of *Clays and Clay Minerals*).

is an index for the transformation, in the vicinity of a basalt intrusion. Recent calculations yield that the heat input will be limited to 10,000 years and temperatures of 100–150°C and is extremely small when compared to that of a basalt intrusion with a temperature of ~1200°C and a heating period of 100 million years. The changes in the values of vitrinite reflectance due to the HLW/SF were found to be extremely small (Jobmann *et al.*, 2013).

Results of extensive experimental studies on microbial activities in clays allow identification of microbial processes which can influence the long-term performance of a deep geological repository for HLW/SF in clay (Meleshyn, 2014). Most important microbial activities are those influencing clay alteration and gas production, which can be expected to be most pronounced in the excavation-damaged zone of host rocks due to increased pore space. Activity of sulfate-reducing bacteria (through production of sulfide) as well as Fe(III)-reducing bacteria (direct reduction of Fe(III) in the clay structure) can negatively influence retention properties of clay as a result of the clay mineral alteration. Along with the other microbial and abiotic gas production processes methane production can lead to an over-pressure and formation of cracks. According to the current state of knowledge (Meleshyn, 2014), methanogen microbes remain active up to 122°C, whereas sulfate-reducing bacteria can survive at such temperatures only in an inactive state. They were proved however to have an optimum growth at 80–95°C. Therefore, heating of the clay in excess of 122°C can lead to a considerable decline of microbial population and of microbial activity in the near field of a repository.

Evaluation of the temperature effects with regard to long-term safety

Evaluation of the thermo-hydro-mechanical effects

Heating of the clay to temperatures below the ECT (50 to 80°C) generally has a slightly negative effect on its mechanical properties. Heating to temperatures above the ECT up to ~150°C, on the contrary, has a positive effect on the thermal conductivity, mechanical strength and stiffness of clay. If the temperature rises above the ECT in the near-field around the waste canisters, irreversible consolidation in that part of the host rocks counterbalances the thermal expansion.

Water permeability increases with temperature up to ~80°C and decreases afterwards up to 150°C.

Heating increases the pressure required for gas penetration into the clay. Water evaporation will only take place temporarily during the re-saturation. Subsequently, the boiling point of pore water will increase to ~280°C due to the pressure increase.

Based on current knowledge, the analysis of the thermo-hydro-mechanical effects reveals no deterioration but does reveal a positive impact of a temperature increase beyond the ECT (50–80°C) up to 150°C on the barrier properties of host rocks.

Evaluation of the chemical-biological-mineralogical effects

The period during which the thermal input of a HLW/SF repository heats the host rocks to temperatures relevant for abiotic chemical and mineralogical processes only within a limited period of time and within a distance limited to several metres around the emplacement boreholes or tunnels. During this time no appreciable abiotic illitization (and thus no change in swelling properties) thermochemical sulfate reduction or kerogen transformation to raw oil and natural gas is to be expected. However, substantial illitization, sulfate reduction and natural gas production can occur at much lower temperatures if driven microbially. A temperature increase above 122°C will lead to a considerable decline of the population and activity of sulfate- and Fe(III)-reducing as well as methane-producing microbial species and thus will potentially improve the repository performance. The positively regarded smectitization of illite possibly occurs within this time, but still to a non-appreciable extent. The influence of the cementation through silica and illite neo-formation on the barrier performance of clay as well as possible changes of sorption properties at temperatures ≤150°C can be judged as negligible too.

Sulfate- and Fe(III)-reducing bacteria can build endospores enhancing their resistance to unfavourable environmental conditions. The lethal temperatures for bacteria in an endospore state are 30–40°C higher than those for bacteria in an active state (Nicholson *et al.*, 2000). The upper temperature limit for the activity of sulfate- and Fe(III)-reducing bacteria is at about 95°C and 122°C respectively. Therefore, a relatively short thermal treatment at ~150°C will be necessary in order to eliminate possible endospores in a repository material. With increasing duration of the thermal treatment, the elimination temperature can be expected to decrease according to the observation of only spurious or even no microbial biomass in

sedimentary rocks exposed to palaeo-temperatures of 140°C or 145°C, respectively, during their diagenesis (Colwell *et al.*, 1997). Summarizing, the surviving microbial population in clay will decrease with increasing temperature and duration of the thermal treatment in excess of 122°C.

Final evaluation and definition of the temperature criterion

The above-discussed possible effects of increased temperatures on the barrier properties of the host rocks are summarized in Table 1. It should be noted here that most studies were carried out at temperatures below ~150°C, so that for higher temperatures no evaluation can be made based on the current state of knowledge. As a result of the

TABLE 1. Evaluation of the thermally induced effects in clay with respect to the impact on its barrier properties (ECT = Expansion-Contraction-Threshold) based on available studies for temperatures from 20°C to 150°C.

No.	Thermally-induced effect	Impact evaluation
	Thermo-hydro-mechanical effects	
1	Expansion/contraction of claystone	$T <$ ECT: negative $T >$ ECT: positive
2	Mechanical properties	$T <$ ECT: negative $T >$ ECT: positive
3	Hydraulic properties	$T <$ ECT: negative $T >$ ECT: positive
4	Thermal properties	$T <$ ECT: negative $T >$ ECT: positive
5	Swelling pressure	Negligible
6	Dehydration of swelling clay minerals	Negligible
7	Generation of hydraulic gradients	Negligible
8	Vapourization	Negligible
9	Gas entry pressure	Negligible
	Chemical, biological, and mineralogical effects	
10	Illitization of smectites	Negligible
11	Smectitization of illites	Positive, while negligible
12	Cementation by generation of silica and new minerals	Negligible
13	Thermo-chemical sulfate reduction	Negligible
14	Sorption properties	Negligible
15	Kerogen transformation	Non existent
16	Microbial activities	$T <$ 122°C: negative $T >$ 122°C: positive

evaluation, two processes can be identified as significant: thermal expansion and contraction from the group of the thermo-hydro-mechanical processes as well as microbial activity from the group of the chemical-biological-mineralogical processes.

The impact of increased temperatures on the expansion and contraction can be concluded to be positive as soon as the temperature in the clay host rock exceeds the ECT and as long as the pore pressure due to the thermal expansion of pore fluid, possibly in combination with the thermally induced contraction of the solids, does not lead to fissuring. Besides, along with the repository fields with $T >$ ECT in the close vicinity of the emplaced HLW/SF there are expected to be zones with increased temperatures below the ECT, where rock properties will be influenced negatively. Such zones are however unavoidable in any design of HLW/SF repositories.

Similarly, the impact of increased temperatures on microbial activity can be concluded to be positive as soon as the temperature in the clay exceeds 122°C by several degrees for several tens or hundreds of years. This would lead not only to a temporal prohibition of the microbial activity able to substantially deteriorate barrier properties of clay, but also to a strong decline of microbial populations in, or even to a temporal "sterilization" of the excavation-damaged zone. Such a sterilization can have a sustainable effect, as a recolonization of this part of the host rock along with its cooling-down would be prevented by its irreversible consolidation at $T >$ ECT.

As a result, the temperature criterion is proposed as follows:

"It has to be demonstrated that a temperature of 150°C will not be exceeded in host rocks".

This temperature limit allows development of a disposal concept that considers the possibility of a sterilized zone around the waste canisters. The limit is set to 150°C in view of the lack of relevant studies and information for higher temperatures.

Conclusions

Besides some slight negative effects of heating clay up to ~80°C, a temperature increase up to 150°C leads, amongst others, to increases in thermal conductivity, mechanical strength, and stiffness of the clay, as well as to limiting microbial activities as soon as the environmental temperature exceeds 122°C by several degrees. Thus, heating clay host

rocks does not necessarily compromise but may actually improve the barrier performance.

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