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On the impact of temperatures up to 200 °C in clay repositories with bentonite engineer barrier systems: A study with coupled thermal, hydrological, chemical, and mechanical modeling

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ABSTRACT

One of the most important design variables for a geological nuclear waste repository is the temperature limit up to which the engineered barrier system (EBS) and the natural geologic environment can be exposed. Up to now, almost all design concepts that involve bentonite-backfilled emplacement tunnels have chosen a maximum allowable temperature of about 100 °C. Such a choice is largely based on the consideration that in clay-based materials illitization and the associated mechanical changes in the bentonite (and perhaps the clay host rock) could affect the barrier attributes of the EBS. However, existing experimental and modeling studies on the occurrence of illitization and related performance impacts are not conclusive, in part because the relevant couplings between the thermal, hydrological, chemical, and mechanical (THMC) processes have not been fully represented in the models. This paper presents a fully coupled THMC simulation of a nuclear waste repository in a clay formation with a bentonite-backfilled EBS for 1000 years. 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.

The model simulations demonstrate some degree of illitization in both the bentonite buffer and the surrounding clay formation. Other chemical alterations include the dissolution of K-feldspar and calcite, and precipitation of quartz, chlorite, and kaolinite. In general, illitization in the bentonite and the clay formation is enhanced at higher temperature. However, the quantity of illitization is affected by many chemical factors and therefore varies a great deal. The most important chemical factors are the concentration of K in the pore water as well as the abundance and dissolution rate of K-feldspar; less important ones are the concentration of sodium and the quartz precipitation rate. In our modeling scenarios, the calculated decrease in smectite volume fraction in bentonite ranges from 1 to 8% of the initial volume fraction of smectite in the 100 °C scenario and 1–27% in the 200 °C scenario. Chemical changes in the 200 °C scenario could also lead to a reduction in swelling stress up to 15–18% whereas those in the 100 °C scenario result in about 14–15% reduction in swelling stress for the base case scenario. Model results also show that the 200 °C scenario results in a much higher total stress than the 100 °C scenario, mostly due to thermal pressurization. While cautions should be taken regarding the model results due to some limitations in the models, the modeling work is illustrative in light of the relative importance of different processes occurring in EBS bentonite and clay formation at higher than 100 °C conditions, and could be of greater use when site specific data are available.

1. Introduction

Geological repositories for disposal of high-level nuclear waste generally rely on a multi-barrier system to isolate radioactive waste from the biosphere. The multi-barrier system typically consists of the natural system (NS), which includes the repository host rock and its surrounding subsurface environment, and the engineered barrier system (EBS), which comprises the waste canister and in many design concepts a smectite clay-based buffer or backfill. Clay/shale formations have been considered as a host rock throughout the world and are being studied at several underground research laboratories, e.g., Toarcian argillites at the Tournemire site, France (Patriarche et al., 2004), Opalinus Clay at the Mont Terri, Switzerland (Bossart et al., 2002), and Boom Clay at the Mol site, Belgium (Barnichon and Volckaert, 2003). An EBS composed of clay-based material such as bentonite is routinely considered in the design of nuclear waste repositories. Both host clay rock and EBS bentonite have low permeability, low diffusion rates, high retention capacity for radionuclides, and the capability of swelling that could seal fractures induced by damage due to tunnel excavation, ventilation, and/or heating.

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Radioactive waste (spent fuel) emanates a significant amount of thermal energy due to decay processes, which causes temperature increases in the surrounding environment particularly in the early stages of waste emplacement. The temperature to which the EBS and natural rock can be exposed is one of the most important design variables for a geological repository, because it determines waste package spacing, distance between disposal galleries, and therefore the overall size (and cost) of repository for a given amount of heat-emanating waste (Horseman and McEwen, 1996). This is especially important for a clay repository, because argillaceous rocks have relatively small heat conductivity. A thermal limit of about 100 °C or lower is imposed unanimously in all disposal concepts throughout the world despite their differences in design concepts (Hicks et al., 2009). Chemical alteration and the subsequent changes in mechanical properties are among the determining factors. A high temperature could result in chemical alteration of buffer and backfill materials (bentonite) within the EBS through illitization and cementation, which compromise the function of these EBS components by reducing their plasticity and capability to swell when wetting and to maintain swelling pressure (Pusch and Karnland, 1996; Pusch et al., 2010; Wersin et al., 2007). The swelling capability of clay minerals within the bentonite is important for sealing gaps between bentonite blocks, between bentonite and other EBS components, and between the EBS and the surrounding host rock, inhibiting the water infiltration into EBS and subsequently the corrosion of canister, and suppressing the microbial activity. The swelling capacity loss due to illitization therefore will compromise these beneficial features of EBS. Chemical alteration may also occur in the near-field host rock, which could reduce the clay capability for self-sealing within the excavation damaged zone (EDZ). As a result of the low permeability of a clay rock, a high temperature may induce significant pore pressure build-up (through pore water expansion and vaporization) in the near field, which could generate adverse mechanical deformation (such as fracturing), damaging the integrity of the host rock (Horseman and McEwen, 1996).

Regarding the concern of chemical alteration and the associated mechanical changes, Wersin et al. (2007), after reviewing a number of data sets, concluded that the criterion of 100 °C for the maximum temperature within the bentonite buffer is overly conservative. Their conclusion was based on their findings that no significant changes in bentonite hydraulic properties occur at temperatures of up to 120 °C under wet conditions and that bentonite is chemically stable to much higher temperature under dry conditions. The impact of a high temperature on bentonite and clay host rock behavior, and the consequences on repository performance, are largely open questions for a clay repository system. While various studies shed light on certain aspects of this question, there is a lack of studies that integrate the relevant THMC processes and consider the interaction between the EBS and host rock. In this paper, we use coupled THMC modeling to evaluate the chemical alteration and mechanical changes in EBS bentonite and the NS clay formation under various scenarios, attempting to provide useful information for decisions on temperature limits and motivate more studies on this issue.

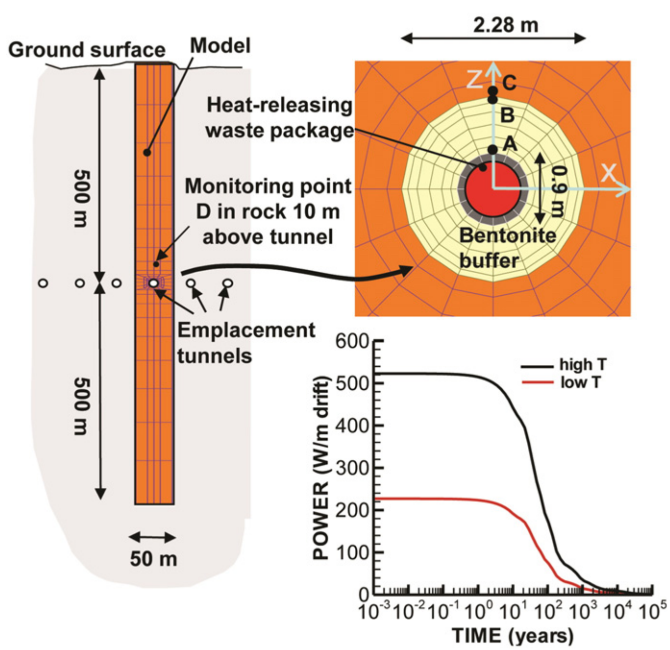

Fig. 1. Domain for the test example of a bentonite back-filled horizontal emplacement drift at 500 m (Rutqvist et al., 2014). Modeling monitoring points: A: inside the bentonite near the canister, B: inside the bentonite and near the EBS-NS interface, C: inside the clay rock formation and near the EBS-NS interface, D: inside the clay rock formation at a distance of 10 m from the canister. "High T": 200 °C; "Low T": 100 °C.

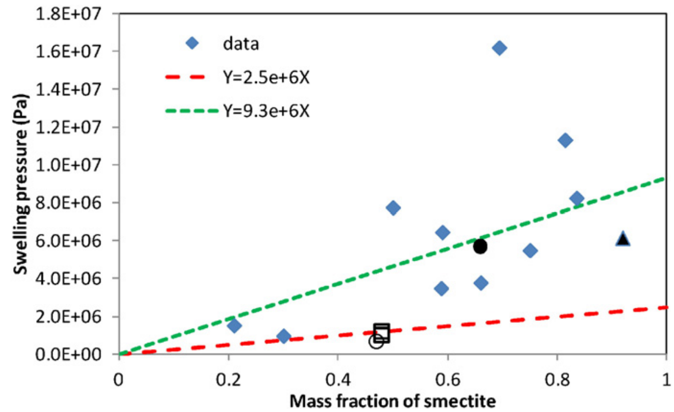

Fig. 2. Swelling pressure versus mass fraction of smectite for various bentonites. ▲, FEBEX bentonite (ENRESA, 2000); ●, Montigel bentonite (Bucher and Müller-Vonmoos, 1989); □, Kunigel VI bentonite (JNC, Japan Nuclear Cycle Development Institute, 1999); ○, Kunigel bentonite (Komine and Ogata, 1996), ♦ are data for reference material from Czech, Danish, Friedland, Milos Deponit CA-N, Kutch (Indian) and Wyoming MX-80 (Karnland et al., 2006). The lines are linear regression curves.

2. A review of the chemical and mechanical alteration in EBS and clay formation

A number of studies have been conducted to evaluate the chemical, mechanical and geological alteration in bentonite and clay-rock formations as a result of hydration and/or heating at temperatures ranging from 30 to 300 °C. Illitization, the transformation of smectite to illite, has caught great attention of researchers because it results in a loss of smectite which in turn causes a loss in the swelling capacity. Illitization is evident in geological systems (Wersin et al., 2007), as exemplified by several natural analog studies (Pusch and Madsen, 1995; Kamei et al., 2005; Cuadros, 2006; Casciello et al., 2011). Illite/smectite mixed-layer clay is commonly observed in clayey sediments, and deep formations

Table 1
Mineral volume fraction (dimensionless, ratio of the volume for a mineral to the total volume of medium) of the EBS bentonite (Ochs et al., 2004) and the clay formation (Bossart, 2011; Lauber et al., 2000) used in the model.

Mineral	Clay formation: opalinus clay	EBS bentonite: Kunigel-V1
Calcite	0.093	0.016
Dolomite	0.050	0.018
Illite	0.273	0.000
Kaolinite	0.186	0.000
Smectite	0.035	0.314
Chlorite	0.076	0.000
Quartz	0.111	0.228
K-Feldspar	0.015	0.029
Siderite	0.020	0.000
Ankerite	0.045	0.000