



Research paper

Hydrogeochemical evolution of the bentonite buffer in a KBS-3 repository for radioactive waste. Reactive transport modelling of the LOT A2 experiment



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ARTICLE INFO

Article history:

Received 8 March 2012

Received in revised form 11 September 2014

Accepted 15 September 2014

Available online 7 October 2014

Keywords:

Bentonite buffer

High level nuclear waste

LOT experiment

Reactive transport modelling

TOUGHREACT

ABSTRACT

The Swedish Nuclear Fuel and Waste Management Company (SKB) is conducting a series of long term tests at the Äspö Hard Rock Laboratory to assess the behaviour of the bentonite buffer under conditions similar to those expected in a KBS-3 repository for high level nuclear waste. The LOT A2 experiment consists of a vertical borehole with a central heater inside a copper tube surrounded by compacted bentonite. During four years, the temperature of the copper tube was maintained at 130 °C, while the bentonite was progressively water saturated by the injection of groundwater. During this period, physical and hydro-geochemical data were collected. By using the code TOUGHREACT, a model was made to simulate the processes of solute transport which control the chemical and the mineralogical distribution observed in the bentonite at the end of the test. Additionally, a series of sensitivity analyses was performed to assess the influence of key parameters controlling the thermal–hydro-geochemical processes. Numerical results indicate that, within the first year, the heated bentonite blocks are completely water saturated, which agree with the measured data. The simulated transport of chloride, the dissolution/precipitation of Ca sulphate minerals, and the cation redistribution in the montmorillonite interlayer also agree with data measured at the end of the experiment.

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1. Introduction

The deep geological repository is the most widely accepted solution for the final deposition of high level nuclear waste (HLNW). In the KBS-3 concept for a deep geological repository, designed by the Swedish Nuclear Fuel and Waste Management Company (Bjergsson et al., 2005; Wikberg, 2012), a compacted bentonite buffer is placed around copper canisters containing spent nuclear fuel, isolating it from the hydrogeological environment. The physical–chemical properties of the compacted bentonite buffer provide mechanical stability, hydrological isolation, and chemical buffering, which provides the retardation of an eventual radionuclide release. In this way, the geochemical stability of the bentonite buffer is essential for the performance of this HLNW repository concept.

Since 1999, SKB is conducting a series of tests in the Long Term buffer material (LOT) at the Äspö Hard Rock Laboratory (HRL) (Karnland et al., 2009), in order to check hypotheses for the evolution of the bentonite buffer under the thermo-hydraulic conditions expected in a KBS-3 repository. Under these conditions, the inner border of the bentonite buffer is heated by the copper canister which induces the

evaporation of the porewater. At the outer border, the saturated host rock induces a hydro-mechanical pressure over the bentonite, leading to its water saturation (Fig. 1, Hökmark, 2004; Villar et al., 2008). During this process, clay minerals take up water, swell and reduce the pore space, leading to the reduction of bentonite permeability (Komine, 2004; Kröhn 2003). During water saturation of the bentonite, advection plays an important role on the transport of solutes. Nevertheless, as soon as the bentonite becomes fully water saturated, the transport of solutes is dominated by diffusion. Depending on the type of solute, other processes such as osmosis and geochemical reactions can also influence solute mobility. Chloride transport, for instance, can be markedly influenced by Donnan equilibrium or osmosis (Keijzer et al., 1999; Bjergsson and Karnland, 2009), while the transport of cations, like calcium, sodium, potassium and magnesium, is also influenced by cation exchange reactions in the montmorillonite interlayer (Arcos et al., 2003; Bjergsson and Karnland, 2009). Finally, ion transport and their distribution along the aqueous phase (e.g., calcium, sulphate and silica) are key processes controlling the dissolution of primary minerals and the precipitation of secondary phases (Arcos et al., 2003; Wersin, 2003).

Due to the important role of the bentonite buffer as a hydro-mechanical and a geochemical barrier, laboratory and in-situ experiments, natural analogue studies (i.e. studies of natural systems with features and processes similar to those expected in a deep geological

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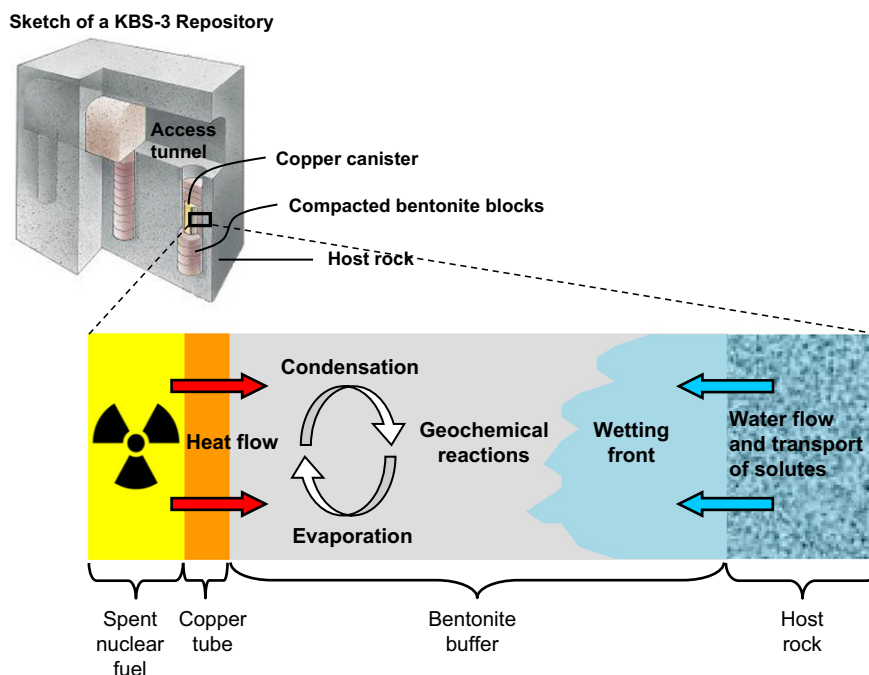


Fig. 1. Sketch of the thermo-hydraulic, geochemical and transport processes that are believed to undergo in the bentonite buffer of a KBS-3 repository.

repository), and numerical models have been developed (Arcos et al., 2003; Wersin, 2003; Hökmark, 2004; Kozaki et al., 2005; Montes et al., 2005; Wersin et al., 2007; Villar et al., 2008; Karnland et al., 2009; Marty et al., 2010; Itälä et al., 2011). In the present work, by simulating the results obtained during the LOT A2 experiment, the potential impact of (1) the thermo-hydraulic processes on the transport of solutes, and (2) the geochemical reactions on the buffer properties of the bentonite are quantitatively evaluated for the first time. The robustness of the numerical models developed here is the consequence of a parameterization that is based on laboratory results (Villar et al., 2008), field-scale experiments (Karnland et al., 2009), and previous modelling exercises (Arcos et al., 2003, 2006). By building such robust models the numerical results provide a solid basis for evaluating the effect of the hydrothermal conditions, which induce porewater evaporation, condensation and suction, on the geochemical behaviour of the bentonite buffer in a KBS-3 deep geological repository for HLNW. The work presented here represents a major step forward with respect to the state-of-the art of the process understanding and numerical simulation of the hydrogeochemical processes that are likely to occur in the bentonite buffer, since it reproduces accurately the analytical data of the bentonite samples collected in the LOT A2 experiment. In addition, an uncertainty analysis has been developed to assess the main parameters controlling the thermal-hydro-chemical processes that influence the transport and retention of solutes in the bentonite buffer.

2. Description of the LOT A2 experiment

2.1. Geometry and thermo-hydraulic conditions

The LOT A2 experiment was installed in a core-drilled borehole in the Äspö Hard Rock Laboratory at a depth of 450 m below ground surface, on October 29, 1999, and lasted six years. It is composed of a central heater surrounded by a copper tube and 100 mm of compacted bentonite placed between the copper tube and the granitic host rock. The heater is intended to reproduce the heat generated by the radioactive decay of high level nuclear waste. Albeit the highest temperature expected for the canisters in a KBS-3 repository is 90 °C, in the LOT A2 experiment the heater was set to 130 °C at the lowest 2 m of the borehole (Karnland et al., 2009), in order to assess the adverse consequences of

higher temperatures. During the first year, the temperature within the bentonite buffer progressively increased reaching a thermal gradient that was stationary during the last five years of the experiment (Fig. 2).

Temperature, total pressure, water pressure and water content were measured during the heating period. The sensors (thermocouples; optical sensors and vibrating wire sensors for total and water pressure; and, Vaisala moisture sensors for water content) were connected to a standard PC-based data acquisition system, and registrations were made every hour. All moisture sensors operating during the LOT A2 experiment indicate a considerably fast increase in humidity to over 90% within the first year, meaning that during most of the experiment the bentonite was almost fully water saturated (Karnland et al., 2009).

2.2. Hydrochemical conditions

Before the ongoing of the LOT A2 experiment, selected bentonite blocks were chosen for physical and hydrogeochemical analyses. The data obtained from the analyses of these reference bentonite blocks represent the initial conditions prior to the LOT A2 experiment. The porewater composition was measured from aqueous leachates which were prepared by dispersion of 6 g of bentonite solid mass with 60 mL of deionised water, and considering a gravimetric water ratio (mass of porewater/mass of dry clay) of 10%, under laboratory conditions, according to Karnland et al. (2009). Conversion from the concentrations of these aqueous leachates to the concentration of ions in the initial porewater of the bentonite (expressed in $\text{mol} \cdot \text{L}^{-1}$) is as follows:

$$[\text{Species}] (\text{mol} \cdot \text{L}^{-1}) = \frac{0.06 \text{ L deionized water}}{6 \text{ g dry clay}} \cdot \frac{1 \text{ g dry clay}}{0.1 \text{ g porewater}} \cdot \frac{1000 \text{ g porewater}}{1 \text{ L porewater}} \quad (1)$$

where $[\text{Species}] (\text{mol} \cdot \text{L}^{-1})$ is the concentration (in $\text{mol} \cdot \text{L}^{-1}$) of a given chemical species in the aqueous leachate.

The initial composition of the bentonite porewater prior to the LOT A2 experiment (Table 1, column 2) was calculated by using Eq. (1), and the results from the aqueous leachate (Table 1, column 1).

The concentration of exchangeable cations like sodium, potassium, calcium, and magnesium, and also the sulphate concentration in the

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