



Reactive transport modelling of groundwater-bentonite interaction: Effects on exchangeable cations in an alternative buffer material in-situ test



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ARTICLE INFO

Article history:

Received 25 February 2016

Received in revised form

8 June 2016

Accepted 15 July 2016

Available online 18 July 2016

Keywords:

Reactive transport modelling

Exchangeable cations

Groundwater interaction

Temperature-dependant diffusion

Bentonites

ABM

In-situ scale

ABSTRACT

Bentonite clays are regarded a promising material for engineered barrier systems for the encapsulation of hazardous wastes because of their low hydraulic permeability, swelling potential, ability to self-seal cracks in contact with water and their high sorption potential. SKB (Svensk Kärnbränslehantering) has been conducting long term field scale experiments on potential buffer materials at the Äspö Hard Rock Laboratory for radioactive waste disposal in Sweden.

The Alternative Buffer Material (ABM) test examined buffer properties of eleven different clay materials under the influence of groundwater and at temperatures reaching up to 135 °C, replicating the heat pulse after waste emplacement. Clay materials were emplaced into holes drilled in fractured granite as compacted rings around a central heater element and subsequently brought into contact with groundwater for 880 days. After test termination, and against expectations, all clay materials were found to have undergone large scale alterations in the cation exchange population. A reactive-diffusive transport model was developed to aid the interpretation of the observed large-scale porewater chemistry changes. It was found, that the interaction between Äspö groundwater and the clay blocks, together with the geochemical nature of the clays (Na vs Ca-dominated clays) exerted the strongest control on the porewater chemistry. A pronounced exchange of Na by Ca was observed and simulated, driven by large Ca concentrations in the contacting groundwater. The model was able to link the porewater alterations to the fracture network in the deposition hole. The speed of alterations was in turn linked to high diffusion coefficients under the applied temperatures, which facilitated the propagation of hydrochemical changes into the clays. With diffusion coefficients increased by up to one order of magnitude at the maximum temperatures, the study was able to demonstrate the importance of considering temperature-dependant diffusion in understanding and predicting geochemical alterations of engineered barriers systems after relatively short exposure times following waste emplacement.

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1. Introduction

Bentonite clays are candidate materials for engineered barrier systems for the encapsulation of hazardous wastes because of their low hydraulic permeability, swelling potential, ability to self-heal cracks in contact with water and their sorption potential (Pusch,

2001). Compacted bentonite buffers are being investigated in deep geological repositories for radioactive waste for their suitability to encapsulate waste canisters and ability to seal access galleries, isolating the nuclear waste from the hydrogeological environment. Accordingly, the geochemical stability of bentonite clays is essential for the performance and safety evaluation of waste repositories.

As part of these safety assessments, experiments on bentonites durability and stability over time and under different thermal, geochemical and mechanical conditions at different scales have been performed over the years. Potential mineral transformations

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and alterations in porewater chemistry and mineralogy under well-defined conditions have been investigated in laboratory experiments in order to predict solute mobility and the geochemical stability of bentonites. During the re-saturation phase after placement of spent nuclear fuel, the transport of solutes in bentonite clays has been found to be controlled by advection (e.g. Salas et al., 2014). However, following saturation, the main transport process is, due to clays' low hydraulic conductivity, by molecular diffusion. In order to predict solute migration under conditions expected in a repository, the diffusive behaviour within compacted bentonites has been investigated in laboratory experiments for different species (e.g. water, anions, cations, colloids) (e.g. Appelo et al., 2010; Gimmi and Kosakowski, 2011; González Sánchez et al., 2008; Melkior et al., 2009; Van Loon et al., 2007 among others); under different temperatures (González Sánchez et al., 2008; Kozaki et al., 1998; Suzuki et al., 2004) and as a function of compaction (Pusch, 2002; Suzuki et al., 2004). Nevertheless, diffusion within bentonites is still under discussion in regards to a mechanistic representation of the underlying phenomena (Idiart and Pekala, 2016). Uncertainties result from a lack of a well-established and widely accepted conceptual model of the structure of compacted bentonite at the nano-scale at different dry densities and salinities. Available characterization techniques for porosity distribution and nano-structure of compacted bentonite often perturb the structure of the water saturated bentonite and are therefore of limited applicability. In addition, the validity of the classical Fickian diffusion approach based on a single bulk (total) porosity has been questioned in some cases, as it is unable to represent the diffusion of cations, anions and neutral species simultaneously. As a result, more complex mechanistic models have been proposed in the literature to explain experimental data, based on different conceptualizations of the pore space (e.g. Birgersson and Karnland, 2009; Appelo et al., 2010; Gimmi and Kosakowski, 2011; Tachi and Yotsuji, 2014).

Solute mobility in compacted bentonites is further influenced by geochemical reactions such as cation and anion exchange (e.g. Valter and Plötze, 2013; Birgersson and Karnland, 2009; Tachi and Yotsuji, 2014), as well as the dissolution of primary minerals or the precipitation of secondary phases (Liu and Neretnieks, 2006; Arcos et al., 2008; Kaufhold et al., 2013; Idiart et al., 2013), which impact on the performance of bentonites as barriers by altering e.g. the swelling capacity or the hydraulic and gas conductivities of these clays. In order to upscale laboratory findings and to test the behaviour of bentonite clays under the thermo-hydraulic conditions expected in radioactive waste repositories, several countries are conducting large scale in-situ tests of engineered barrier systems. Villar and Lloret (2007) investigated thermo-hydro-mechanical properties of a Spanish bentonite after five years treatment at repository conditions in the so-called FEBEX experiment in Grimsel, Switzerland. Aertsens et al. (2013) compared the results of tracer migration experiments in the Boom Clay Formation in the underground research laboratory in Mol in Belgium with laboratory experiments. For more than a decade, Swedish Nuclear Fuel and Waste Management Co. SKB (Svensk Kärnbränslehantering) has been conducting long term field scale experiments at the Äspö Hard Rock Laboratory in Sweden. The "Long Term Test of Buffer Material" project (LOT; Olsson and Karnland, 2011; Karnland et al. 2000; Karnland et al. 2009) focused on in-situ thermo-hydraulic processes and their impact on solute transport and buffer properties of MX-80 bentonite. In 2006 another field scale experiment was launched, the Alternative Buffer Material (ABM) test, (Eng et al., 2007; Svensson et al., 2011), which extended investigations to a range of clays, including compacted sedimentary clay materials (Friedland clay, Germany and Callovo-Oxfordian clay, France) and various compacted bentonites. Three

columns consisting of these different compacted clay buffer blocks were installed in three boreholes (ABM1–3). The analysis of the first package, ABM1, is the subject of this paper. The aim of the ABM1 test was to compare different clay materials and their performance over time when in contact with the heater, the seals, the fractured host rock, and ambient groundwater at relevant depths as well as the expected maximum in-situ temperatures. The end goal is thereby to provide a basis for selecting an ideal buffer for the Swedish spent nuclear fuel repository. The experiment consists of a vertical borehole placed at –450 m depth (Svensson et al., 2011). It contains a central heater inside an iron tube surrounded by 11 different compacted clay blocks. The column of blocks was artificially saturated with Äspö groundwater and heated to a maximum temperature of 130 °C. Over a duration of 28 months data were recorded on the temperature and water pressure evolution. Following the test, a comprehensive characterization of the cation exchange population and distribution with time (Dohrmann et al., 2013) as well as mineralogical alterations (Kaufhold et al., 2013) was carried out. All buffers started with distinctly different exchangeable cation (EC) populations and it was expected that, after the field experiment, the ECs would display horizontal variations for all bentonite rings due to the radial heat distribution induced by the central heater element. However, in contrast to this, large scale variations in the ion exchange composition had occurred. That is, the composition of exchangeable cations had changed markedly in all clay materials over the whole test parcel after 28 months, but no significant horizontal variations in the EC populations within each ring was detectable. The most significant change was an increase in exchangeable Ca^{2+} , and a decrease in exchangeable Na^+ and Mg^{2+} . Mineralogical changes of the bentonites were unable to explain these significant changes, and it was postulated that interaction with the surrounding groundwater must have caused partial equilibration of the porewater chemistry within and possibly between the different bentonite blocks within a short time frame (Idiart et al., 2014; Dohrmann et al., 2013; Pekala et al., 2012).

In order to assess this hypothesis and to enhance our understanding of the changes in porewater chemistry in bentonites under the influence of in-situ groundwater and heat conditions, a reactive geochemical model of the ABM1 field test was developed. In this study the aim was (i) to develop a numerical model that provides a process-based description of the physical and geochemical mechanisms controlling the chemical evolution of bentonite porewaters at the field (i.e. meter) scale; (ii) to quantify the influence of groundwater on the chemical evolution of the different clays with time, and (iii) to verify the model by applying it to the available field data set collected during the ABM1 test.

2. Materials and methods

2.1. ABM1 set-up

The set-up of the ABM1 experiment, which is described in detail in Eng et al. (2007) and Svensson et al. (2011), consists of a central steel canister with heaters, surrounded by ring shaped bentonite blocks situated in a borehole of 300 mm diameter in crystalline bedrock at about 450 m depth (Fig. 1). Eleven different clay materials (two sedimentary clays and nine bentonites) were included in the tests as compacted rings (Table 1 and Fig. 1) with an outer diameter of 280 mm, and an inner diameter of 100 mm. Additionally, four rings were included which consisted of cloth-covered steel-cages filled with bentonite pellets with and without additional quartz to also test granulated material. All clay materials were installed multiple times and the total number of material blocks stacked on top of each other was 30 (Fig. 1). Each block had a

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