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Research paper

# Thermal–hydraulic–mechanical behavior of bentonite and sand-bentonite materials as seal for a nuclear waste repository: Numerical simulation of column experiments

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## ARTICLE INFO

## Article history:

Received 20 May 2016

Received in revised form 3 October 2016

Accepted 4 October 2016

Available online xxxx

## Keywords:

High-level radioactive waste

Buffer material

Column experiments

Numerical model

THM simulation

## ABSTRACT

Depending on the repository concept, buffer materials around the canister may play an important role for the performance of deep geological repositories for high-level radioactive waste (HLW). Coupled thermo-hydro-mechanical (THM) processes are initiated in the buffer material, as heat produced by the HLW typically causes an initial drying of this sealing layer, which may be followed by re-saturation with fluid from the host rock. In this study, a fully coupled and process-oriented numerical model was applied to simulate laboratory experiments investigating two buffer materials, i.e. clay pellets as well as a sand-bentonite mixture. The developed models account for heat transport, multiphase flow and mechanical effects from swelling and thermal expansion. Model calibration was achieved by first determining the most sensitive parameters for heat and multiphase flow effects. A good fit between experimental data and model results was achieved for temperature, relative humidity, water intake and swelling pressure measurements. It is found that a unique set of thermal parameters for the insulation materials of the two experiments can be identified, which allows for a reliable estimate of the heat balance of the experiments. The most important parameters identified are the intrinsic and relative permeability, the water retention curve and the saturation dependent thermal conductivity of the buffer materials. It is further shown that the transient saturation state of the buffer material strongly influences its hydraulic and thermal behavior. The buffer material behavior thus identified contributes to the evaluation of larger scale in-situ experiments, and to better design deep geological repositories.

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## 1. Introduction

Depending on the repository concept, the design of deep geological repositories for high-level radioactive waste (HLW) may require the presence of a sealing engineered barrier between the waste canisters and the host rock in order to slow down the transport of radionuclides, prevent the formation of flow paths to the host rock, and contribute to the mechanical stability of the tunnel. The behavior of a HLW repository is characterized by the heat produced through radioactive decay of the radionuclides in spent fuel and vitrified high level waste. In the early phase of the post closure period, the temperature near the canister may reach 140 °C and desaturation is expected in the buffer. Simultaneously, water may circulate from the host rock to the buffer material, transporting dissolved ions from the canister and also causing changes in the chemistry of the material and affecting its mechanical behavior (Espina Gomez and Villar, 2010). Therefore, the thermal conductivity of the buffer material must be sufficient to prevent too high

temperatures in the near field of the canister (Posiva, 2006; Rautioaho and Tantu, 2009), while the hydraulic permeability must be low enough to prevent water circulation. Bentonite has been considered as a suitable sealing material because of its low permeability, swelling capacity and high retention properties but its low thermal conductivity at dry conditions might lead to high temperatures within the buffer and the possibility of shrinkage might allow water circulation (Alonso et al., 2005; Cui et al., 2000; Pintado and Lloret, 2006). In order to increase the thermal conductivity and reduce the shrinkage, mixtures of bentonite with varying percentages of sand have been investigated (Arnedo et al., 2008; Cho et al., 2011; Liu et al., 2014; Ruedi et al., 2013).

The feasibility of a deep geological repository in respect to temperature changes is evaluated with tests performed both at the field and laboratory scale in order to identify the thermo-hydro-mechanical (THM) processes taking place in the engineered barriers and the surrounding host rock (Villar et al., 2012). Within the PEBS project ([www.pebs-eu.de](http://www.pebs-eu.de)) a 1:2 scale in-situ experiment, termed HE-E, is carried out at the underground laboratory in Mont Terri, Switzerland, to improve the understanding of the thermal evolution of the near field around a HLW canister during the early phase after emplacement (Czaikowski et al.,

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2012; Gaus et al., 2014a, 2014b). The experiment consists of two tunnel sections with heaters representing the waste canisters. One section is filled using pure MX-80 bentonite pellets, while the other section is filled with a mixture of 35% MX-80 and 65% quartz, having a higher thermal conductivity (Villar et al., 2014).

For characterizing the buffer materials, well controlled laboratory THM experiments of the MX-80 and the sand-bentonite mixture were conducted (Villar et al., 2012, 2014), mimicking in the laboratory the expected field conditions of heating and hydration. The two column experiments were performed at the Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas (CIEMAT) as a part of the PEBS Project. These experiments were chosen as a task of the DECOVALEX (DEvelopment of COupled models and their VALidation against Experiments; [www.decovalex.org](http://www.decovalex.org)) international cooperative research project phase VI (2012–2015). The aim of DECOVALEX is to develop mathematical models and quantify coupled THM processes in fractured rocks and buffer/backfill materials by simulation of well controlled experiments and by comparison of the model results from the individual participating groups (Hudson and Jing, 2013; Jing et al., 1995).

Our work presented here is a part of the DECOVALEX project and focuses on the numerical simulation of the two above mentioned laboratory column experiments. Our modelling work aims to identify and include the relevant processes influencing the thermal–hydro–mechanical behavior of the tested materials, identify the sensitive parameters and thus quantify the relevant material behavior.

These aims are pursued by calibration of the model to the available measurements for temperature, humidity and water intake using the same set of thermal parameters for the insulation set-up of both experimental columns, thus only varying the properties of the MX-80 and the sand–bentonite mixture.

This modelling effort contributes to providing reliable quantitative parameters for the two buffer materials, which can then be applied for the in-situ field-scale HE-E experiment.

## 2. Materials and methods

### 2.1. Filling material: MX-80 bentonite pellets and sand-bentonite mixture

The materials tested in the column experiments (PEBS project) are the same as those used as buffer in the two sections of the field HE-E experiment (Villar et al., 2012, 2014) and were parameterized in this study by using numerical simulations. The grain size distributions of the MX-80 and the sand–bentonite mixture are reported in Fig. 1a (Villar et al., 2012, 2014; Rizzi et al., 2012) but the MX-80 granulometric curve is apparent as the material used came in the form of pellets of

different sizes at nominal dry densities of  $1.53 \text{ g cm}^{-3}$  and a saturation of 22% (Villar et al., 2014). The studied MX-80 bentonite was formed in hydrothermal alteration of volcanic ash during the Cretaceous period. An x-ray diffraction test performed by the University of Lausanne and reported in Rizzi et al. (2012) indicates that the bentonite is composed by about 70% of montmorillonite with one water layer (exchangeable cations are Na, K and/or Ca), 15% quartz, and 10% feldspar (plagioclase and FEK). The pore size distribution of the bentonite pellets was determined by Villar (2013) using mercury intrusion porosimetry (Fig. 1b), showing a predominant pore mode at about  $0.014 \mu\text{m}$  (mesopore) and a second peak at  $470 \mu\text{m}$  (macropore).

The sand-bentonite mixture is constituted of 65% quartz sand (diameter ranging from 0.5 to 1.8 mm) and 35% Na-bentonite GELCLAY WH2 (granular material of the same composition as MX-80) of the same grain spectrum (Villar et al., 2012). As shown in Fig. 1b (modified from Villar, 2013), the mixture has a predominant macroporosity with a pore mode at about  $204 \mu\text{m}$  and a second mode at  $0.02 \mu\text{m}$  (mesopore). The nominal dry density of the mixture is  $1.45 \text{ g cm}^{-3}$  with an initial saturation of 11% in order to reproduce the setting expected in the field experiment (Villar et al., 2014).

Although sand has a higher thermal conductivity than bentonite, Wieczorek et al. (2011, 2013) tested the sand–bentonite mixture, obtaining values of thermal conductivity similar to those of the MX-80 bentonite. The same authors suggested that the reduced specimen sizes used may have influenced the results, as confirmed by the fact that when determining the water retention curve of the material, suction was already close to zero at 28% saturation. The swelling pressure of the mixture was measured in standard oedometers and values ranging from 1.5 to 0.7 MPa were obtained (Villar et al., 2012).

### 2.2. Experimental set-up

The THM behavior and the properties of MX-80 bentonite and the sand-bentonite mixture were experimentally investigated within the PEBS project in two column experiments reproducing the expected conditions in the deep geological HLW repository of the current Swiss concept in the early post-closure phases (Villar et al., 2012, 2014). The experimental set-up is described in detail in Villar et al. (2012, 2014). In the following, the main characteristics of both cells are given in order to facilitate the understanding of the conceptual and numerical models developed in our work. In order to reproduce the heating of the buffer material and the re-saturation caused by the water recirculation from the host rock, the experiments were divided into 1) a heating phase and 2) a heating and hydration phase. Heat was applied in the experiments by a heater positioned at the bottom of the columns. The heating phase is characterized by a stepwise increase of the

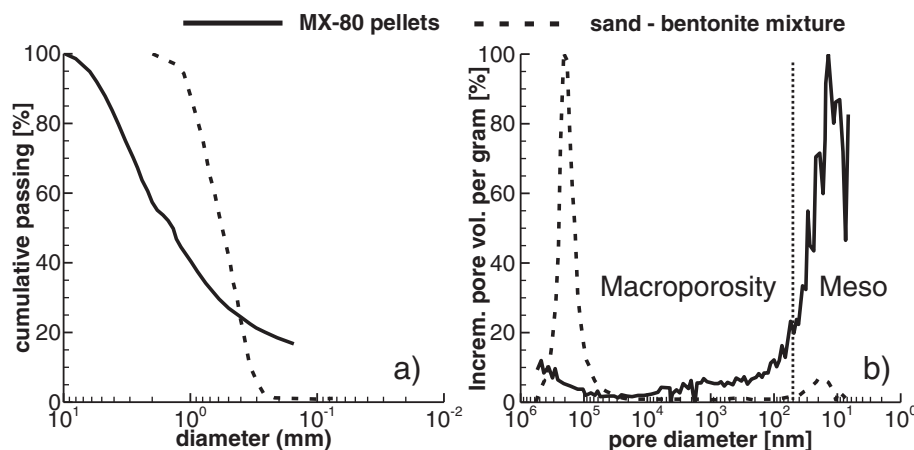


Fig. 1. a) granulometric curve; b) pore size distribution of the bentonite pellets and sand-bentonite mixture. (Modified from Villar, 2013)

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