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Research paper

Comparison of the hydrogeochemical and mechanical behaviours of compacted bentonite using different conceptual approaches

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ABSTRACT

This study compares the capabilities of two numerical models that simulate the behaviour of compacted saturated MX-80 bentonite exposed to different chemical environments. Both codes have different approaches: one of them is a reactive transport model, and the other is a hydrogeomechanical model. The study shows that the combined use of both approaches provides a useful basis for designing bentonite barriers in the geological disposal of high-level nuclear waste. Although, their application is now limited to simple geometries and boundary conditions, the proposed approaches are a first step towards a fully thermo-hydro-mechano-chemical model. It is also shown that there is a need for a greater understanding of the chemomechanical coupling that occurs at the microstructural scale to achieve this comprehensive model in the future.

1. Introduction

Bentonite is the most promising candidate buffer and backfill material for in engineered barrier systems in subsurface repositories of high-level nuclear waste. The current design in Sweden and Finland is called KBS-3 and has two alternatives, KBS-3V (SKB, 2010; Posiva Oy, 2012) and KBS-3H (Posiva Oy, 2013), both using bentonite as barrier. This natural material has favourable properties such as its swelling capacity, its low permeability at full saturation, and its capacity to passivate or retard certain geochemical processes (Arthur and Zhou, 2000; Arthur et al., 2005). Consequently, there is an objective need for understanding the complex behaviour of this material in the short, medium and long term (SKB, 2010). For example, the swelling pressure developed by a bentonite buffer is an important design consideration (SKB, 2010; Posiva Oy, 2012). According to safety requirements, swelling pressures should lie within a range of 2–15 MPa (Posiva Oy, 2012). Values higher than this may damage the canister and therefore significantly affect the safety of the repository. Variations in the swelling pressure may be due to multiple factors (Raiko et al., 2010), including the geometry and arrangement of the buffer with respect to the fractures of the host rock, the mineralogy of the bentonite, the type of predominant counterions (e.g., Na^+, Ca^{2+}, Mg^{2+}), or the chemical composition of the surrounding groundwaters. It has been verified experimentally that the chemistry of the saturating water affects the permeability (Dixon, 2000; Cho et al., 2002; Karnland et al., 2006; Castellanos et al., 2008; Zhu et al., 2013), the strength properties (Di Maio and Fenelli, 1994; Börgesson et al., 1995; Di Maio, 1996; Jeong et al., 2012; Sinnathamby et al., 2015), the deformational properties (Bolt, 1956; Mesri and Olson, 1971; Sridharan and Rao, 1973; Low, 1980; Di Maio et al., 2004; Calvello et al., 2005; Castellanos et al., 2006; Ye et al., 2014; Zhu et al., 2015; Chen et al., 2016) or the mineralogical composition due to processes such as illitization (Hökmark et al., 1997; Wersin et al., 2007) or chloritization, among other processes (Karnland and Birgersson, 2006; Gómez-Espina and Villar, 2010; Marty et al., 2010; Xiaodong et al., 2011; Gómez-Espina and Villar, 2015). However, the swelling pressure under different salinity conditions and types of solutes has traditionally been the most experimentally studied variable (Low, 1980; Pusch, 1980; Dixon et al., 1996; Karnland, 1998; Dixon, 2000; Karnland et al., 2006). In all cases, an increase in the ionic strength of the saturating fluid translates into a reduction of the swelling pressure (Savage, 2005). Consequently, it is essential to understand the long-term chemical composition of the surrounding groundwater of a repository. However, even though a detailed experimental characterization of the material (Herbert et al., 2004; Karnland et al., 2005, 2006; Yamaguchi et al., 2007; Herbert et al., 2008) and its behaviour with different pore water compositions has already been performed, the slowness of the processes, which is associated with the system's low saturated hydraulic conductivity, does not allow for real-scale

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experimentation in many cases. This type of in situ testing has only been performed in a few studies (Karnland et al., 2000; Alonso et al., 2005; Pacovský et al., 2007; Svoboda, 2007; Li et al., 2013; Martín et al., 2014; Liu et al., 2016) with long test times, high costs, and variable degrees of interpretation and understanding.

In this context, the numerical modelling of the multiphysics processes that occur in bentonite is important. Historically, in a first approach, mechanical processes were coupled with hydraulic (i.e., HM models) (Alonso et al., 1990; Gens and Alonso, 1992; Börgesson et al., 1996) and thermal processes (i.e., THM models) (Olivella et al., 1996; Thomas and He, 1997; Navarro and Alonso, 2000). It is less common to find multicomponent geochemical transport models in conjunction with those previously cited (i.e., THMC models) because the coupling of all these phenomena is complicated (Guimarães et al., 2013; Lei et al., 2014; Sedighi et al., 2015). Additionally, there is no single conceptual model that combines all of these processes effectively and accurately. Thus, different approaches have been used to reduce the complexity of the chemomechanical coupling, either by reducing the amount of chemical species and reactions involved in the geochemical model or by simplifying the mechanical constitutive model. This study evaluates the ability of two models, each of which uses one of the aforementioned simplifications, to estimate the evolution of different variables, both chemical and mechanical, and also evaluates their capability to be used in a complementary manner in the design of engineered barrier systems for spent nuclear fuel and other nuclear wastes. Furthermore, this study is a first step before a complete conceptual and numerical thermo-hydro-mechano-chemical model, for which a better understanding of the phenomena involved is still needed.

2. Description of the models

The first of the numerical models considered in this study is the CRUNCHFLOW model (Steefel et al., 2015), which is a geochemical transport model with a simplified auxiliary geomechanical model implemented in the CRUNCHFLOWMC version; this model is referred to as the mC model in this study. The second model, developed in a Comsol Multiphysics environment, was proposed by Navarro et al. (2017), which is primarily mechanical with a simplified geochemical model; this model is referred to as the Mc model in this study. Both models use a double-porosity approach to describe the compacted bentonite system. In this study, macroporosity is defined as the aggregate pore space, and microporosity is defined as the intra aggregate pore space (Romero et al., 2011). Both models also define chemical equilibrium between ions in the two porosity domains using the Donnan equilibrium approach (Tournassat and Appelo, 2011; Alt-Epping et al., 2015). The Donnan equilibrium substitutes cation exchange: the total positive charge in the microporosity, balancing the negative charge of the clay mineral layer, represents the cation exchange capacity of the bentonite. The composition in the microporosity reflects the exchangeable cation occupancy. The Donnan pore space in both models is considered to be the total microporosity; the presence of a Stern layer containing immobile cations near the negatively charged montmorillonite sheet surface is not explicitly considered. Any montmorillonite edge site reactions have been discarded; no significant pH changes are expected in this numerical experiment. Therefore, the potential buffer role that edge sites can play via protonation or deprotonation will not be relevant in this comparison. The ability to simulate the flow of the liquid phase in unsaturated conditions is also a common feature in the two models, although only problems full saturation will be analysed in this study. Finally, solute transport occurs in both microporosity (diffusive only) and in macroporosity (both advective and diffusive). Diffusion coefficients can be set individually for micro- and macroporosity.

Despite the common conceptual structures, ionic distribution and solute transport methods of the models, there are important differences in the modelling approaches with regard to geochemical modelling. The mC model is a multicomponent reactive transport code including thermodynamic equilibria, kinetic mineral precipitation/dissolution reactions, surface complexation, and conventional ion exchange (Alt-Epping et al., 2015; Steefel et al., 2015). On the other hand, the geochemical system of the Mc model is limited because it only considers one anion (Cl^-) and two cations (Na^+ and Ca^{2+}) and does not consider any reactions between them. Although this chemical system can reproduce behaviour trends that have been observed in various tests in the literature (Karnland et al., 2006; Castellanos et al., 2008; Ye et al., 2015), the chemical phenomena that alter the original mineralogy of bentonite (Karnland and Birgersson, 2006) cannot be simulated by these models.

In terms of geomechanical modelling, the levels of sophistication of both codes are opposite. In the Mc model, a complete elastoplastic constitutive model (Navarro et al., 2013, 2014) was chosen based on the Barcelona Expansive Model (BExM) (Alonso et al., 1999; Sánchez et al., 2005), although it includes differentiating elements. First, the model Mc includes a formulation of the water exchange kinetics between micro- and macroporosities that considers the chemical potential difference between both porosity levels (Navarro et al., 2016). The model also uses a constitutive model of the behaviour of the microstructure that is based on the interpretation of retention curves at high suction values (Navarro et al., 2015) and incorporates an alternative definition of the interaction function between the micro- and macrostructural strains to consider the free swelling phenomena and the effect of the salinity of the saturating water (Navarro et al., 2017). The result is a model that includes the major parameters controlling the hydromechanical behaviour of the bentonite.

In the mC model, the microporosity volume is calculated from the Debye length, which is inversely proportional to the square root of ionic strength in the macroporosity (Tournassat and Appelo, 2011). Because the Debye length overlaps in compacted bentonites, the interlayer thickness is a fraction of Debye length, and this parameter (termed multiplier) is considered as constant for fixed bentonite dry density. Microporosity volume can then be derived by multiplying interlayer thickness with the total specific surface area of montmorillonite. Macroporosity follows from the total volume constraint and the known dry density. The Debye length multiplier is derived from experimental data stating interlayer distances in MX80 bentonite at different dry densities and known macroporosity ionic strength (Holmboe et al., 2012). The final expression for the calculation of the microporosity (n_m) is

$$n_\mathrm{m} = SSA \cdot \lambda \cdot \frac{\beta}{\sqrt{I}} \tag{1}$$

where *SSA* is the total specific surface area of montmorillonite, β is a time-dependent physical constant (3.03743×10^{-10} m, Tournassat and Appelo, 2011), λ is the multiplier. Moreover, there is no geomechanical module to solve the mechanical equilibrium equation. An empirical correlation proposed by the authors is used to apply experimental data found in the literature (Karnland et al., 2006). The expression

$$P_{swell} = 2 \cdot b \cdot \mathrm{arcoth}(a \cdot I + d) + c \tag{2}$$

links ionic strength I (M) and swelling pressure P_{swell} (kPa), the parameters in Table 1 result in satisfactory fits of the experimental data for both sodium- and calcium-dominated bentonites (Fig. 1) for a

Table 1
Parameters from fitting Eq. (2) to measured data (Fig. 1).

Parameter	Na-montmorillonite	Ca-montmorillonite
a	111.8	24.8
b	510,672.4	600,913.8
c	−460.1	−1613.4
d	215.2	257.8