



Research paper

Model evaluation of geochemically induced swelling/shrinkage in argillaceous formations for nuclear waste disposal



Liange Zheng^{*}, Jonny Rutqvist, Hui-Hai Liu, Jens T. Birkholzer, Eric Sonnenthal

Berkeley National Laboratory (LBNL), Berkeley, CA 94720, USA

ARTICLE INFO

Article history:

Received 4 October 2013

Received in revised form 7 May 2014

Accepted 16 May 2014

Available online 18 June 2014

Keywords:

Argillaceous

Swelling

Diffuse double layer

Geochemistry

Nuclear waste disposal

ABSTRACT

Argillaceous formations are being considered as host rocks for geologic disposal of nuclear waste in a number of countries. One advantage of emplacing nuclear waste in such formations is the potential self-sealing capability of clay due to swelling, which is of particular importance for the sealing and healing of disturbed rock zones (DRZ). It is therefore necessary to understand and be able to predict the changes in swelling properties within clay rock near the waste-emplacement tunnel. In this paper, considering that the clay rock formation is mostly under saturated conditions and the swelling property changes are mostly due to geochemical changes, we propose a modeling method that links a THC simulator with a swelling module that is based on diffuse double layer theory. Simulations were conducted to evaluate the geochemically induced changes in the swelling properties of the clay rock. Our findings are as follows: (1) geochemically induced swelling/shrinkage occurs exclusively in the EBS–clay formation interface, within a few meters from the waste-emplacement tunnels; (2) swelling/shrinkage-induced porosity changes are generally much smaller than those caused by mineral precipitation/dissolution processes; (3) geochemically induced swelling/shrinkage of the host clay rock is affected by variations in the pore water chemistry, exchangeable cations, and smectite abundance. Neglecting any of these three factors might lead to a miscalculation of the geochemically induced swelling pressure.

© 2014 Elsevier B.V. All rights reserved.

1. Introduction

Argillaceous formations are primary host-rock candidates for nuclear waste disposal, and have therefore been studied extensively, especially in Europe. Extensive studies on the behavior of clays have been conducted in Underground Research Laboratories (URLs), such as in Opalinus Clay at Mont Terri, Switzerland (Bossart and Thury, 2008; Pearson et al., 2003; Thury, 2002), and in Callovo-Oxfordian Clay at Bure, France (Andra, 2005; Jougnot et al., 2010; Samper et al., 2008). Research and development efforts for shale repository investigations were also undertaken in the U.S. from the 1970s until the mid-1980s (Gonzales and Johnson, 1984; Hansen et al., 2010), before the U.S. nuclear waste program was directed to exclusively characterize the volcanic tuffs at Yucca Mountain in Nevada. In fact, the U.S. has many clay/shale/argillite basins with positive attributes for permanent disposal of nuclear waste. Recently, Hansen et al. (2010) presented a feasibility study, indicating that shale formations provide a technically advanced, scientifically sound disposal option for the U.S.

One advantage of emplacing nuclear waste in clay formations is the potential self-sealing capability of clay due to swelling. The self-sealing capability might have particular importance for disturbed rock zones (DRZ). These are zones that form near emplacement tunnels as a result

of excavation and tunnel disturbance. It is, therefore, necessary to evaluate and be able to predict the changes in swelling properties within clay rock, especially in the near-field area. In the long run, the clay rock in the near field area stays in a saturated condition although it undergoes desaturation in the short term, the swelling-property changes in a clay rock are therefore mostly due to the geochemical changes, including (1) changes in ion concentration of the pore water, (2) cation exchange changes in the composition of the water in the interlayer space, and (3) changes in the abundance of swelling minerals such as smectite. Geochemically induced changes in the swelling properties of clay rock have been reported in both short-term and small-scale laboratory tests (Thury, 2002; Wakim et al., 2009). Thury (2002) reported that decompressed Opalinus Clay swells in contact with water and that the magnitude of swelling varied with the water chemistry (deionized water, low mineralized water, synthetic pore water, and KCl solution). Wakim et al. (2009) experimentally evaluated the effect of aqueous solution chemistry on the swelling and shrinkage of shales under saturated conditions, and reported that more concentrated solution led to less swelling. Kamei et al. (2005) and Cuadros (2006) indicated that illitization, i.e., the transformation from smectite to illite, changes the swelling properties of the clay. Although whether illitization would occur in a nuclear waste disposal site and to what degree it happens is still an issue that requires more studies, the changes in the swelling properties of clay rock demonstrated in these studies point out the need to evaluate geochemically induced swelling/shrinkage within the

^{*} Corresponding author.

E-mail address: lzheng@lbl.gov (L. Zheng).

clay rock hosting a nuclear waste repository, and to the best of our knowledge, no such evaluation has been reported in the literature. It has been recognized that the self-sealing of DRZ fractures could lower the risk brought by the DRZ (Tsang et al., 2010) and it is largely determined by the swelling of the clay minerals (Rothfuchs et al., 2007). Although attention has been paid to self-sealing due to hydraulic effect (Zhang, 2011), the effect of geochemical changes on the swelling and the subsequent self-healing of clayey rock has not been evaluated.

In this paper, after linking the thermal, hydrological and chemical (THC) simulator TOUGHREACT (Xu et al., 2011) with a swelling model based on the Gouy–Chapman diffuse double layer (DDL) theory, generic simulations were conducted to evaluate the geochemically induced changes in the swelling properties of clayey host rock in the near field of a nuclear waste repository.

2. The simulator—TOUGHREACT

TOUGHREACT is a numerical simulator for chemically reactive nonisothermal flows of multiphase fluids in porous and fractured media (Sonnenenthal et al., 2005; Spycher et al., 2003; Xu and Pruess, 2001; Xu et al., 2006, 2011; Zhang et al., 2008; Zheng et al., 2009). The code was developed by introducing reactive chemistry into the multiphase fluid and heat flow simulator TOUGH2 (Pruess, 2004). The code accommodates any number of chemical species present in liquid, gas, and solid phases and considers a variety of subsurface thermal, physical, chemical, and biological processes, under a wide range of pressure, temperature, water saturation, ionic strength, pH and Eh conditions. The major chemical reactions that can be considered in TOUGHREACT include aqueous complexation, acid–base, redox, gas dissolution/exsolution, cation exchange, mineral dissolution/precipitation, and surface complexation.

3. Application of DDL theory to calculate swelling pressure

In expansive clay–water–ion systems, there are basically two types of swelling processes: crystalline and osmotic. Crystalline swelling refers to swelling that occurs at relatively low water content and is primarily a consequence of the hydration of interlayer exchangeable cations. Swelling occurs as water enters the mineral interlayer as a sequence of successive molecular layers, which results in a step-wise separation of the interlayer for up to three or four layers of water. Osmotic swelling occurs at higher water contents and is associated with continuing interlayer separation that develops from movement of water into the interlayer due to the difference in ion concentration within the interlayer and within the bulk pore water. Osmotic water adsorption results from concentration differences among dissolved ions between the interlayer pore water (overlapping double layers) and the free (bulk) water. It is a long-range interaction, which mostly depends on ionic strength or ion concentration, the type of exchangeable ion (e.g., Ca vs. Na), pH of the pore water, and clay mineralogy (van Olphen, 1977). Corresponding osmotic swelling results from the balance of attractive and repulsive forces that develop between overlapping electrical double layers. Crystalline swelling, which occurs mainly in the initial hydration, will be transformed to osmotic swelling when the clay approaches full saturation. Wayllace (2008) reported that such a transformation will occur when the relative humidity (RH) is around 97%, and Onikata et al. (1999) indicated, from a microscopic point of view, that the $d(001)$ -value of 20 to 40 Å is probably a critical value, where the electrostatic attractive force between the 2:1 layers by way of the cations is so weak that the crystalline swelling is transformed into osmotic swelling.

The swelling of clays can be modeled in several ways. Elastoplastic models (Gens and Alonso, 1992; Thomas and He, 1998), such as the Barcelona Basic Model (BBM) (Alonso et al., 1990) and the Barcelona Expansive Model (BExM) (Alonso et al., 1999) are widely used. Recently, chemical components are also incorporated into the stress–strain

relation, such as incorporation of the concentration of exchangeable cations (Guimarães et al., 2007) and consideration of the aqueous concentration via chemical potential (Ghassemi and Diek, 2003). Another approach involves a constitutive equation to relate the deformation of compacted bentonite to the distance between two montmorillonite layers based on the DDL theory (e.g., Komine and Ogata, 1996; 2003; Schanz and Tripathy, 2009).

Based on the fact that: (1) the host argillaceous formations stay fully saturated despite a short desaturation period in the near field and, (2) the driving force is mostly the geochemical changes, the clay rocks will undergo osmotic swelling when in contact with the engineered barrier system (EBS). Typically, the argillaceous host rock has a pore water ion concentration higher than the bentonite in the EBS, which implies that near the interface, ions will diffuse between the host rock and the EBS bentonite, which will induce a disequilibrium of chemical potential and therefore osmotic swelling. This will occur depending on the degree of compaction of the bentonite and if changes in dry densities are allowed.

During the osmotic swelling process, the swelling pressure is the difference between the osmotic pressure in the central plane between two clay plates and the osmotic pressure in the equilibrium solution according to the DDL theory (Bolt, 1956). In other words, the swelling pressure is the pressure required to keep the clay–water system at the required void ratio when it is allowed to adsorb water or electrolytes (Tripathy et al., 2004). Bolt (1956) and van Olphen (1977) presented a method for calculating the swelling pressure in a clay–water electrolyte system. Sridharan and Jayadeva (1982) improved that method and presented the diffuse double layer theory in a lucid form that could be readily used for understanding the engineering behavior of clays. According to Sridharan and Jayadeva (1982), swelling pressure is determined through a combination of Eqs. (1) to (5) as follows:

$$e = G\gamma_w Sd \quad (1)$$

where, e is the void ratio, G is the specific gravity of soil solids, γ_w is the unit weight of water, S is the specific surface area of soil (m^2/g swelling clay), and d is half the distance between parallel clay platelets.

$$\int_z^u \frac{1}{\sqrt{(2 \cosh y - 2 \cosh u)}} dy = \int_0^d d\xi = -Kd \quad (2)$$

where, u is the nondimensional midplane potential, z is the nondimensional potential at the clay surface, y is the nondimensional potential at distance x from the clay surface, and ξ is the distance function ($=Kx$). K ($1/\text{m}$) is the double layer parameter:

$$K = \sqrt{\frac{2n(e')^2 v^2}{\epsilon kT}} \quad (3)$$

where, e' is the elementary electric charge ($1.6 \cdot 10^{-19}$ C), k is Boltzmann's constant ($1.3806 \cdot 10^{-23}$ J/K), n is the molar concentration of ions in pore fluid (molal), v is the valence of the interlayer cation, T is the absolute temperature (K), ϵ is the dielectric constant of the pore fluid and given by $\epsilon = \epsilon_0 D$ in which ϵ_0 is the permittivity of the vacuum ($8.8542 \cdot 10^{-12}$ C² J⁻¹ m⁻¹), and D is the ratio of the electrostatic capacity of condenser plates separated by the given material to that of the same condenser with vacuum between the plates (Mitchell and Soga, 2005).

$$\begin{aligned} -\left(\frac{dy}{d\xi}\right)_{x=0} &= \sqrt{(2 \cosh z - 2 \cosh u)} \\ &= \Gamma \sqrt{\left(\frac{1}{2\epsilon n k T}\right)} \quad \text{at } x = 0, y = z \\ &= \left(\frac{B}{S}\right) \sqrt{\left(\frac{1}{2\epsilon n k T}\right)} \end{aligned} \quad (4)$$

Download English Version:

<https://daneshyari.com/en/article/1694652>

Download Persian Version:

<https://daneshyari.com/article/1694652>

[Daneshyari.com](https://daneshyari.com)