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Review

Thermo-hydro-mechanical behavior of clay rock for deep geological disposal of high-level radioactive waste

Chun-Liang Zhang

Gesellschaft für Anlagen- und Reaktorsicherheit (GRS), Repository Safety Research Division, Theodor-Heuss-Str. 4, Braunschweig, 38122, Germany

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ABSTRACT

In the context of deep geological disposal of radioactive waste in clay formations, the thermo-hydro-mechanical (THM) behavior of the indurated Callovo-Oxfordian and Opalinus clay rocks has been extensively investigated in our laboratory under repository relevant conditions: (1) rock stress covering the range from the lithostatic state to redistributed levels after excavation; (2) variation of the humidity in the openings due to ventilation as well as hydraulic drained and undrained boundary conditions; (3) gas generation from corrosion of metallic components within repositories; and (4) thermal loading from high-level radioactive waste up to the designed maximum temperature of 90 °C and even beyond to 150 °C. Various important aspects concerning the long-term barrier functions of the clay host rocks have been studied: (1) fundamental concept for effective stress in the porous clay-water system; (2) stress-driven deformation and damage as well as resulting permeability changes; (3) moisture influences on mechanical properties; (4) self-sealing of fractures under mechanical load and swelling/slaking of clay minerals upon water uptake; (5) gas migration in fractured and resealed claystones; and (6) thermal impact on the hydro-mechanical behavior and properties. Major findings from the investigations are summarized in this paper.

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1. Introduction

Clay rocks are world-widely investigated for deep geological disposal of radioactive waste due to their favorable properties such as large homogeneous rock mass, stable geological structure, extremely low hydraulic conductivity, self-sealing potential of fractures, and high sorption capacity for retardation of radionuclides. Several countries have proposed disposal concepts on the basis of a multi-barrier-system, which comprises the natural geological formations and engineered barriers. In France and Switzerland, for instance, the potential repositories for the disposal of high-level radioactive waste (HLW) will be constructed in the indurated Callovo-Oxfordian (COX) and Opalinus (OPA) argillaceous formations, respectively (Nagra, 2002, 2010, 2014; Andra, 2005, 2015).

In the French concept (Andra, 2005, 2015), the repository will be constructed in the COX clay formation at a depth of about 500 m

below the ground surface. HLW canisters will be disposed in horizontal boreholes of 70 cm diameter and tens of meters in length. The boreholes are steel-lined to support the surrounding rock and to ensure emplacement and potential retrieval of waste packages. Each disposal borehole is sealed with swelling clay. The thermal load from HLW is designed to be limited below 90 °C in the rock mass.

In the Swiss concept (Nagra, 2002, 2010, 2014), the repository will be constructed in the OPA formation at a depth of 400–700 m below the ground surface. The facility will include a series of dead-end emplacement tunnels for HLW disposal, which will be excavated in diameter of 2.5–3 m and lengths of several hundred meters. HLW canisters will be emplaced in the middle of the tunnel section and the rest space will be backfilled with expansive bentonite to retard radionuclide migration. The temperature will increase up to 140 °C–160 °C in the buffer on the canister surface and 75 °C–95 °C at the buffer/rock interface.

Construction and operation of a repository will inevitably disturb the rock mass, particularly in the near-field, yielding complex transient thermo-hydro-mechanical (THM) processes and interactions in the natural and engineered barriers for long periods of time of thousands of years. The following disturbances are crucial for the repository safety:

E-mail address: chun-liang.zhang@grs.de.

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- (1) Excavation results in a concentration of the rock stress and redistribution of the pore water pressure, which usually generates an excavation damaged zone (EDZ) around the openings that may act as potential pathways for fluid flow and radionuclide migration into the biosphere.
- (2) Support of the openings restricts extension of the EDZ and ensures the operation safety; however, the support efficiency tends to decrease with time due to alternation and damage of the support materials (e.g. steel and concrete) in chemical interactions with pore water solutions.
- (3) Ventilation with relatively dry air induces suction and causes release of some pore water out of the rock, which may lead to shrinkage of the pore space and increase the inherent cohesion and friction resistance between particles and hence the strength.
- (4) Backfilling and sealing of the repository with suitable materials limit access of groundwater to the waste and thus release of radionuclides via drifts and shafts on one hand, and support the EDZ and limit new damages in host rock on the other hand.
- (5) Heat transfer from HLW produces high temperatures in the near-field, which affects the rock stress and deformation, pore water pressure and transport, chemical interactions between pore water, solutes and mineral surfaces, and causes some changes of the barrier properties of the host rock.
- (6) Gas generation from corrosion of metallic components may cause gas overpressure if the gas generation rate exceeds the gas dissipation rate in the geological and engineered barrier system, which in turn may generate cracks or dilatational pathways for transport of contaminated water and radionuclides to the aquifer and the biosphere.

Assessment of the long-term performance of a clay host rock and thus the safety of the repository needs comprehensive knowledge of its THM properties and responses to the dynamic conditions (Yu et al., 2014; Bernier et al., 2017; Jobmann et al., 2017). During the last two decades, the THM behavior of the clay rocks has been extensively investigated in the underground research laboratories (URLs) in the COX clay rock at Meuse/Haute-Marne in France (MHM-URL) (Armand et al., 2017) and in the OPA at Mont-Terri in Switzerland (MT-URL) (Bossart et al., 2017). A number of large/full-scale experiments have been conducted in the URLs under realistic repository conditions. The in situ experiments are supported by laboratory tests on samples for providing robust database and for understanding the material behavior. On the basis of the test data, constitutive models and computing codes have been developed for prediction of coupled THM processes in the geological and engineered barriers. From the research activities, comprehensive results have been achieved and published mostly in the Proceedings of the International Conferences on Clays in Natural and Engineered Barriers for Radioactive Waste Confinement (Reims 2002; Tours 2005; Lille 2007; Nantes 2010; Montpellier 2012; Brussels 2015; and Davos 2017). Some of the publications dealing with the THM behavior of clay rocks are listed as follows: Noynaert (2000), Zhang et al. (2007, 2010a, 2013), Bock et al. (2010), Tsang et al. (2012), Li (2013), Menaceur et al. (2015, 2016a), Armand et al. (2017), Bossart et al. (2017), Chen et al. (2017) and Marschall et al. (2017).

Within the framework of the German site-independent research and development program, GRS (Gesellschaft für Anlagen-und Reaktorsicherheit) has participated in the international research projects in the URLs and investigated the THM behavior of both COX and OPA claystones with various important aspects concerning the barrier functions of the clay host rocks, including:

- Conceptual stress model for clay rock;
- Stress-driven deformation, damage, reconsolidation and permeability changes;

- Moisture influences on the mechanical properties and behaviors;
- Self-sealing of fractures;
- Gas migration in fractured and resealed claystones; and
- Thermal effects on the hydro-mechanical behaviors and properties.

This paper presents the most important findings from our investigations. Details can be found in the respective references.

2. A conceptual stress model

The studied clay rocks are a complex material in terms of mineralogical composition, microstructural organization and state of pore water (Yven et al., 2007; Mazurek et al., 2008; NEA Clay Club, 2011; Robinet et al., 2015; Menaceur et al., 2016b). Fig. 1 illustrates schematically the microstructure and the state of pore water in the COX claystone, which is similar for the OPA claystone. The pore sizes in claystones mainly range from nano-scale (<2 nm) in between the parallel platelets of the clay particles to micro- and meso-scale (2–50 nm) between solid particles. The claystones have a clayey matrix embedding other mineral particles (quartz, calcite, and others). The clayey matrix consists of clay particles with strongly adsorbed interlayer water within the sheet structures and with strongly to weakly adsorbed water at the external surfaces. Large pores are filled with water that can freely migrate.

Taking into account the microstructure and the state of pore water, effective stress in claystone has been examined by Horseman et al. (1996), Rodwell et al. (1999) and Zhang (2017a). It is recognized that the effective stress in a water-saturated claystone is partly or even fully transferred by the bound pore water within the interlayer pores and narrow interparticle pores between clay particles. A conceptual stress model was derived by Zhang (2017a), which suggested that the effective stress in a dense clay-water system is transferred through both the solid-solid contact between non-clay mineral grains and the bound water in narrow pores between clay particles. In the model, clay particles including interlayer water are taken as microstructural units since the water molecules in the interlayer are strongly adsorbed and immobile under usually encountered pressure gradients. Fig. 2 shows schematically the stress components acting in any wavy surface that passes through the contact areas between particles in a water-saturated claystone.

The total stress acting on the medium σ_t can be expressed as

$$\sigma_t = \sigma_s + \sigma_1 + p_w \quad (1)$$

where σ_s is the contact stress between solid particles, σ_1 is the average disjoining (swelling) pressure acting in the bound water between clay particles, and p_w is the pressure acting in free water in macropores. Thus the interparticle or effective stress σ_{eff} consists of two parts acting at solid-solid contact area and in interparticle bound water, i.e.

$$\sigma_{\text{eff}} = \sigma_t - p_w = \sigma_s + \sigma_1 \quad (2)$$

In clay-rich and less cemented materials, the effect of solid-solid contacts between non-clay particles disappears, i.e. $\sigma_s \rightarrow 0$, so that the effective stress is mostly carried by the bound pore water and is equivalent to the swelling pressure, i.e. $\sigma_{\text{eff}} \leftrightarrow \sigma_1$. Conversely, if a claystone contains a large number of non-clay particles and/or is strongly cemented, the effect of bound water is negligible, i.e. $\sigma_1 \rightarrow 0$, and thus the externally applied load will be transferred through the solid-solid grain contacts, i.e. $\sigma_{\text{eff}} \leftrightarrow \sigma_s$. This concept provides a reasonable view to the nature of the effective stress in

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