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Full Length Article

Thermal effects on clay rocks for deep disposal of high-level radioactive waste

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A B S T R A C T

Thermal effects on the Callovo-Oxfordian and Opalinus clay rocks for hosting high-level radioactive waste were comprehensively investigated with laboratory and in situ experiments under repository relevant conditions: (1) stresses covering the range from the initial lithostatic state to redistributed levels after excavation, (2) hydraulic drained and undrained boundaries, and (3) heating from ambient temperature up to 90 °C–120 °C and a subsequent cooling phase. The laboratory experiments were performed on normal-sized and large hollow cylindrical samples in various respects of thermal expansion and contraction, thermally-induced pore water pressure, temperature influences on deformation and strength, thermal impacts on swelling, fracture sealing and permeability. The laboratory results obtained from the samples are consistent with the in situ observations during heating experiments in the underground research laboratories at Bure and Mont-Terri. Even though the claystones showed significant responses to thermal loading, no negative effects on their favorable barrier properties were observed.

1. Introduction

Clay rocks are world-widely investigated for deep geological disposal of high-level radioactive waste (HLW) due to their favorable properties such as large homogeneous rock mass, stable geological structure, extremely low hydraulic conductivity, swelling and creep capabilities, self-sealing potential of fractures, and especially high sorption capacity for retardation of radionuclides. In France and Switzerland, for instance, the potential repositories will be constructed in the highly-consolidated Callovo-Oxfordian (COX) and Opalinus (OPA) argillaceous formations, respectively. They will be located at depths of more than 500 m below the ground surface. HLW canisters will be disposed in horizontal steel-cased boreholes according to the French concept (Andra, 2005), and/or emplaced in horizontal drifts and backfilled with bentonite in the Swiss concept (Nagra, 2002).

The clay host rocks will be impacted by thermal load from heat-emitting HLW. The most concern is whether and how the favorable barrier properties of the clay rocks will be altered during the thermal period over several thousands of years under designed temperatures below 90 °C. This important issue has being intensively investigated with laboratory experiments, field observations and theoretical analysis. During the last decade, a number of large- and full-scale heating experiments have been conducted in the underground research laboratories (URLs) at Mont-Terri in Switzerland and Bure in France. For instance, the French National Radioactive Waste Management Agency (Andra) and the German Repository Safety Research Institution (GRS) jointly performed a heating experiment (HE-D experiment) at the Mont-Terri-URL from 2003 to 2005 (Wileveau and Rothfuchs, 2003; Zhang et al., 2007). The in situ experiment was accompanied by laboratory characterization of the thermo-hydro-mechanical (THM) behavior of the OPA claystone and numerical modeling of the coupled THM process in the surrounding rocks. Two other similar heating experiments named TER and TED were sequentially conducted by Andra at the Bure-URL from 2006 to 2012 (Wileveau et al., 2007; Conil et al., 2012). GRS contributed to the TER and TED experiments with laboratory experiments on COX samples to investigate thermal effects on the clay rocks (Zhang et al., 2010, 2013). The thermal experiments were performed not only on normal-sized core samples but also on large hollow cylindrical samples. The tests on normal samples aimed at characterizing thermal effects on deformation

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and strength, pore water pressure, swelling capability, sealing and permeability of fractured claystones, while the large hollow cylinder tests focused on examining thermal impact on the excavation damaged zone (EDZ) around HLW boreholes. Major findings from the laboratory experiments are compared with the field observations during the in situ heating experiments.

2. Characteristics of the claystones

The argillaceous formations are results of a specific geological history that lasted for hundreds of millions of years, beginning with deposition and aggregation of fine-grained particles in seawater, followed by sedimentation and consolidation with expelling of pore water, development of diagenetic bonds between mineral particles, and other processes (Horseman et al., 1996; Bock et al., 2010). They have been highly consolidated to low porosities of 14%–18% and extremely low permeabilities of 10^{-20}–10^{-21} m^2. Fig. 1 illustrates schematically the typical microstructure and the state of pore water in the COX and OPA claystones according to Andra (2005) and Bock et al. (2010). The COX claystone contains 25%–55% clay minerals, 20%–38% carbonates and 20%–30% quartz, while the OPA claystone has higher clay contents (58%–76%), less carbonates (6%–24%) and quartz (5%–28%). The pore sizes mainly range from nanoscale (<2 nm) in between the parallel platelets of the clay particles to micro- and meso-scale (2–50 nm) between solid particles. The fraction of the pores smaller than 20 nm is about 60% in the COX claystone, while approximately 75% of the pores in the OPA claystone have a diameter in the range of 1–25 nm. The fraction of macropores (>50 nm) amounts to less than 10% for the claystones. The claystone matrix contains accessory minerals but mainly clay, which consists of particles with strongly adsorbed interlayer water and strongly to weakly adsorbed water at the external surfaces. Only a small amount of mobile water exists in macropores. Obviously, the neighboring clay particles are not directly connected, but mainly through the bound water-films. The interparticle bound water-films are capable of bearing effective stress (Zhang, 2017). The knowledge of the microstructure and the pore water state in the claystones is helpful for understanding their macroscopic responses to thermal loading and coupled THM processes.

For the laboratory experiments, a large number of core samples were extracted from the in situ test fields. Normal samples were prepared to sizes of diameter/length (D/L) = 50 mm/100 mm and 100 mm/200 mm. Large hollow cylinders were prepared to sizes of 280 mm in diameter and 460–530 mm in length with a central borehole of 100 mm in diameter (d). The initial physical properties obtained from some of the tested COX samples are summarized in Tables 1 and 2. The porosities ranged from 14% to 17%. The measured water contents of 5.5%–7.1% correspond to the saturation degrees of 82%–99%. The other COX and OPA samples used for the thermal experiments showed the similar properties (Zhang et al., 2007, 2010, 2013). It should be pointed out that the samples were unavoidably disturbed by coring and preparation. In order to minimize effects of sampling-induced microfissures, they were hydrostatically recompressed before thermal testing to the in situ lithostatic rock stresses or even higher stresses.

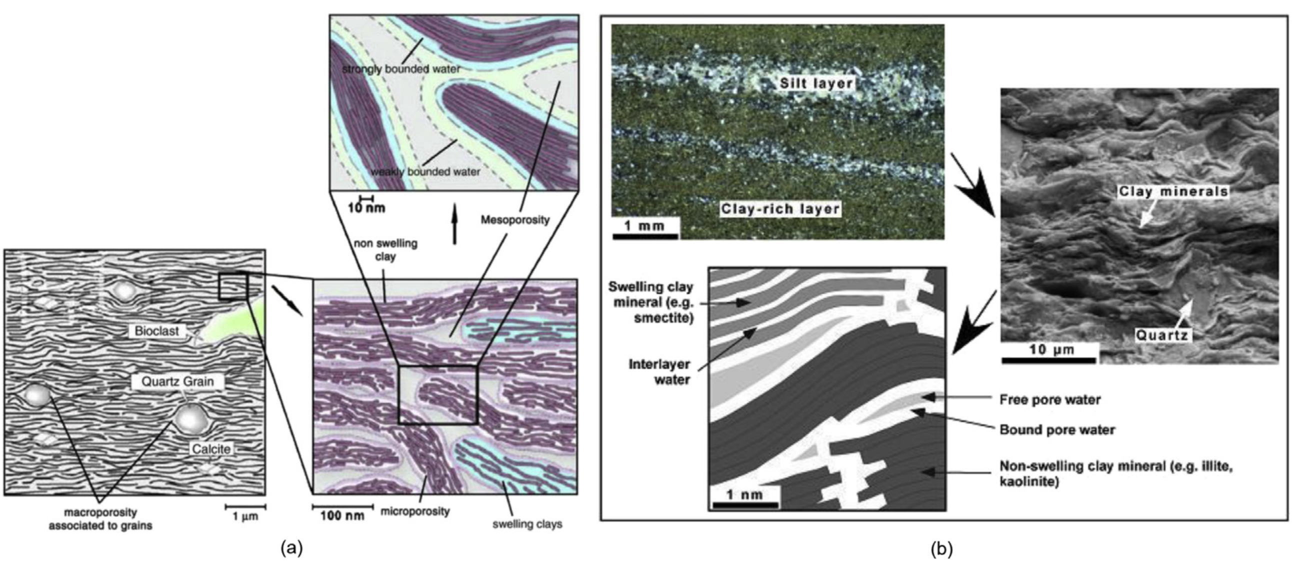

Fig. 1. Schematic sketches of microstructure of (a) the COX and (b) OPA claystones (taken from Bock et al. (2010)).

Table 1
Basic characteristics of some normal samples for thermal testing.

Sample	D/L (mm)	Grain density (g/cm^3)	Bulk density (g/cm^3)	Dry density (g/cm^3)	Porosity (%)	Water content (%)	Water saturation (%)
EST34676	100/200	2.7	2.41	2.24	16.7	7.1	96
EST34678	100/190	2.7	2.42	2.28	15.6	5.9	90
EST33204A	50/100	2.7	2.41	2.26	16.1	6	85
EST33204B	50/100	2.7	2.4	2.26	16.4	6	83
EST33213A	50/100	2.7	2.42	2.28	15.7	6.1	88
EST33213B	50/100	2.7	2.47	2.33	13.8	5.9	99
EST33211A	50/100	2.7	2.45	2.31	14.4	5.7	92
EST33211B	50/100	2.7	2.46	2.32	14.2	5.8	94