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Time- and density-dependent microstructure features of compacted bentonite

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Abstract

Pre-compacted bentonite bricks are often considered as sealing/backfill elements in deep geological repositories for high level radioactive waste. A good understanding of their microstructure changes upon hydration is essential as the microstructure changes are directly related to the macroscopic hydro-mechanical behaviour. In this study, the microstructure features of the compacted MX80 bentonite used as a sealing material in a field experiment were characterized by means of both mercury intrusion porosimetry (MIP) and scanning electron microscopy (SEM). Emphasis was put on the effects of final dry density (density after swelling) and hydration time. The results obtained show that the changes in soil porosity upon swelling are mainly due to the increase in large-pores of about 50 μm diameter and medium-pores of 1 μm diameter. In addition, the microstructure changed over time due to the water re-distribution that occurred among each level of pores: the volume of both the large-pores and small-pores decreased along as the volume of the medium-pores increased. A uniform microstructure can be then expected in the long term. Furthermore, it was observed that the higher the final dry density, the slower the microstructure changes.

Keywords: Radioactive waste disposal; Compacted bentonite; Microstructure; Final dry density; Hydration time

1. Introduction

In the concept of deep geological repository for high-level radioactive waste (HLW), compacted bentonite is often considered as the buffer and sealing material. During its long lifetime, this material will be subjected to thermo-hydro-mechanical (THM) loadings, and it is therefore important, for the safety of the storage system, to well understand its behaviour under such complex and coupled loadings. On the other hand, as the thermo-hydro-mechanical (THM) behaviour of bentonite is strongly dependent on its microstructure changes (Cui et al., 2002; Delage, 2007; Romero and Simms, 2008), a good understanding of its microstructure changes is essential.

Bentonites are mainly composed of smectites, which are made up of structural units (known as laminae or layers) that pile up forming primary particles (Villar et al., 2012); the particles of the clays aggregate together and form aggregates (Delage, 2006). The organisation of the laminae, particles and aggregates gives rise to the following different types of pores (Stepkowska, 1990): (1) inter-layer or inter-laminar pore, which is usually smaller than

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2 nm, (2) inter-particle pore and (3) inter-aggregate pore (larger than 0.002 μm). The inter-layer or inter-laminar pores and inter-particle pores are also known as micro-pores, and the inter-aggregate pores can be divided into macro-pores (> 0.05 μm) and meso-pores (< 0.05 μm) (Villar et al., 2012).

The microstructure feature of bentonite has been qualitatively and quantitatively investigated using scanning electron microscopy (SEM) and mercury intrusion porosimetry (MIP) (Delage, 1996, 2007). SEM is employed to qualitatively visualize the arrangement of particles/aggregates, while MIP is normally used to identify the pore size distribution (PSD), relating the volume of intruded pores to the pressure required for intrusion (Romero and Simms, 2008). Due to the limited pressure range of the MIP technique (up to 200 MPa, depending on the working pressure of the apparatus used), a significant pore volume (entrance diameter smaller than 6 nm) within the aggregates is not detectable (Delage et al., 2006; Lloret et al., 2003; Lloret and Villar, 2007; Nowamooz and Masrouri, 2010; Wang et al., 2013). Transmission electronic microscope or X-ray diffraction (Ben Rhaïem et al., 1985; Saiyouri et al., 1998) can be used to deal with these small pores. Thus, the PSD curve obtained by MIP corresponds to the macro-porosity (> 0.05 μm) and part of the meso-porosity (2 nm to 0.05 μm) according to the definition above. A typical bi-model porosity is usually observed by MIP on compacted unsaturated bentonite (Ahmed et al., 1974; Delage et al., 1996; Romero et al., 1999; Cui et al., 2002; Delage, 2007), defining the small-pores (meso-pores) and large-pores (macro-pores). An increase of dry density reduces the large-pores quantity (Delage and Graham, 1995; Sridharan et al., 1971; Wang, 2012), but does not change the small-pores. Upon hydration, Cui et al. (2002) observed that wetting by decreasing suctions under constant volume condition leads to deformation of aggregates; as a result, the large-pores are progressively clogged.

The microstructure of compacted bentonite can also change over time. Indeed, Delage et al. (2006) investigated the time-dependent microstructure changes of the MX80 bentonite compacted at various dry densities and water contents, and they observed that under the constant volume condition, the microstructure of soil changed over time because of the equilibration of water potential between different types of pores. Their work was however limited to the as-compacted state which corresponds to an unsaturated state. To the authors' knowledge, there has been no investigation into the microstructure changes over different hydration times when the soil samples are fully saturated, even though this corresponds to the situation in a real repository system for nuclear waste.

This effect of hydration time on microstructure changes was investigated in the present work. The soil studied is the pre-compacted MX80 bentonite which is used for the hydraulic seal in the PRACLAY in-situ experiment performed in the underground laboratory of Mol, Belgium. It includes mainly three tests as shown in Fig. 1: the Gallery & Crossing Test, the Heater Test, and the Seal Test. The Heater Test, in which a 30-m long gallery section will be heated for 10 years, aims at investigating a large scale thermal impact of HLW repository on the host Boom Clay. To simulate the most critical state and phenomena in terms of THM responses that could occur within Boom Clay, a low-permeability boundary condition at the head of heater is needed (Tang et al., 2008; Li et al., 2010). For this purpose, an annular seal composed of compacted bentonite was installed between the heated zone and the access gallery (Seal Test in Fig. 1). The bentonite bricks are expected to swell upon hydration, filling the initial technological voids (which have been estimated at 6.9% after installation) and compensating for the deformation of the host Boom Clay. Thus, the dry density of the bricks decreases as a result of swelling. From a practical point of view, it is important to understand the microstructure changes due to the density evolution.

In this study, the microstructure features of MX80 bentonite were investigated by means of both mercury intrusion porosimetry (MIP) and scanning electron microscopy (SEM). To study the density effect, saturated samples with different final dry densities (density after swelling) were prepared by hydrating samples with different values of annular voids. To study the hydration time effect, samples of different dry densities were maintained for various time periods after saturation, and then subjected to microstructure investigation.

2. Materials and methods

2.1. Materials

Samples used in this study were cut from the pre-compacted MX80 bentonite blocks identical to those used for the construction of the hydraulic seal in the PRACLAY test (Seal Test in Fig. 1). The initial dry density of the material was 1.80 Mg/m^3 and the initial water content was 15.22%. The MX80 bentonite

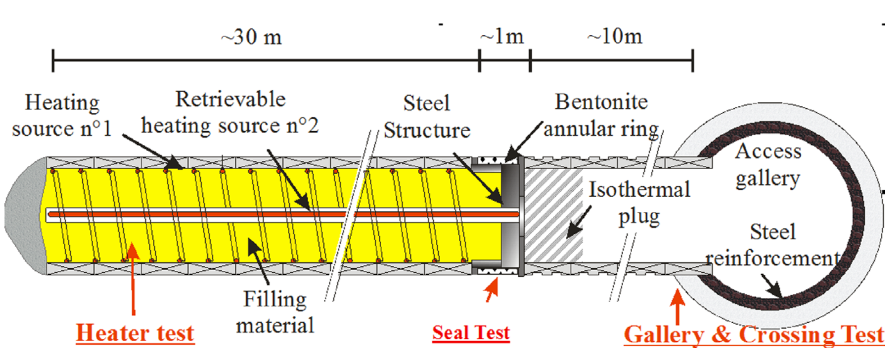

Fig. 1. Overview of the PRACLAY experiment.