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Note

Analysis of the structural changes of a pellet/powder bentonite mixture upon wetting by X-ray computed microtomography

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ABSTRACT

Pellet/powder bentonite mixture is one of the candidate materials for sealing plugs in deep geological high-level radioactive waste disposal. This note presents an investigation on the structure changes of this mixture occurring during the saturation process by means of X-ray computed micro-tomography. The test was performed in an infiltration column (60 mm in inner diameter and 120 mm in height). Water was supplied to the two ends of the column and the changes of the sample morphology were observed during a period of 100 days of hydration. Digital Volume Correlation (DVC) technique was used to determine the vertical displacement field of the bentonite powder. A pressure transducer was used to measure the axial swelling pressure during the hydration. The results show that the initial distribution of powder in the inter-pellet pores was not homogeneous; the powder filled almost completely the pores in the zones close to the two ends while air-filled inter-pellet voids were observed in the middle of the column specimen. When water started to infiltrate inside the specimen from both ends, the pellets and the powder grains started to swell (because of the swelling properties of smectite, the principal mineral of bentonite) and filled the voids. That induced at the same time increase of swelling pressure and downward movement of powder grains. The results allowed a better understanding on the hydro-mechanical couplings, at the pellet scale, in the pellet/powder bentonite mixture upon wetting.

1. Introduction

In the context of deep geological high-level radioactive waste disposal, non-compacted pellet/powder bentonite mixtures are considered as candidate sealing materials because of their high swelling capacity and high radionuclide migration retardation properties, as well as their operational advantages: they are easy to transport, install, and allow gaps between the rock and the seal to be minimized. In a real repository this unsaturated mixture will be installed to plug galleries. Then, water coming from the host rock will start saturating the mixture under almost constant-volume conditions. The swollen mixture will fill different voids and generate swelling pressure, ensuring the sealing of the storage system. Molinero-Guerra et al. (2017) used X-ray computed tomography (X-ray CT) to investigate the microstructure of pellet/powder bentonite and found that this mixture was characterized by an initial heterogeneous distribution of powder grains inside the inter-pellet pores. This initial state could greatly influence the saturation process and the corresponding structure changes. As a result, the overall hydro-mechanical behavior would change during the saturation process. Therefore, to properly assess the safety of the underground radioactive waste disposal, it is of paramount importance to well understand the microstructure change of the mixture in the course of hydration.

In this context, the Institute of Radioprotection and Nuclear Safety (IRSN, France), as a part of the overall research and development program that aims at providing scientific background for disposal safety, launched the SEALEX (SEALing performance EXperiments) project, within which this work was conducted. It consists of a series of full-scale experiments carried out in IRSN's Underground Research Laboratory (URL – Tournemire, France) (Barnichon and Deleruyelle, 2009; Mokni and Barnichon, 2016) and small-scale experiments conducted in the laboratory (Wang et al., 2012; Saba et al., 2014). One of the aims of the full-scale experiments was to test the long-term hydraulic performance of sealing systems in normal conditions for different clay core compositions (pure bentonite or bentonite/sand

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mixtures) and conditionings (pre-compacted blocks, mixture of pellets/powder, or in-situ compacted). In the present work, a pellet/powder bentonite mixture with a proportion of 80/20 in dry mass was investigated.

The hydro-mechanical behavior of engineered barriers composed of compacted bentonite/sand mixtures or compacted pure bentonite has been extensively investigated (e.g. Pusch, 1982; Graham et al., 1989; Komine and Ogata, 1994; Dixon et al., 1996; Alonso et al., 2005; Wang et al., 2013b, 2014; Saba et al., 2014; Sun et al., 2014). However, few studies have been carried out on the mixture of pellet and powder of bentonite. Imbert and Villar (2006) performed infiltration tests on a pellet/powder bentonite mixture and found that the swelling pressure was analogous to that of a specimen of compacted powder at the same dry density. Garcia-Siñeriz et al. (2015) studied the homogenization process of a material made of bentonite pellets through a large-scale experiment on engineered barrier (EB). Upon dismantling after 12 years of operation, a gradient of density and water content was still found within the mixture because of the initial heterogeneity of the material. Van Geet et al. (2005) investigated the hydration process of a pellet/powder bentonite mixture by X-ray CT. A progressive decrease of the pellets density and an apparent homogenous sample after saturation were observed. However, in the work of Van Geet et al. (2005), a cylindrical cell containing only few pellets was used. As a result, the analyses mainly focused on the scale of a single pellet.

The present work was thus conducted to investigate the structural changes of a pellet/powder bentonite mixture with a large number of pellets by X-ray CT. Firstly, the sealing capacity of the mixture and its structural evolution during saturation were studied. Secondly, Digital Volume Correlation (DVC) technique was applied to calculate the displacement field within the sample. Finally, the evolution of the axial swelling pressure while wetting was investigated. The obtained results provide helpful elements to assess the hydromechanical performance of seals/plugs made up of this kind of bentonite mixtures.

2. Material and methodology

2.1. Pellet/powder MX80 bentonite mixture

The soil studied was a mixture of pellet/powder MX80 bentonite (from Wyoming, USA) at a proportion of 80/20 in dry mass. It was provided by the Laviosa-MPC Company under the commercial name Expangel SP7 for pellets and SP30 for the powder, both were produced from the same bentonite. The bentonite had a smectite content of 80%, other minerals being quartz, calcite and pyrite. The CEC was 98 meq/100 g and the major exchangeable cations were: sodium (52 cmol(+)/kg), calcium (37 cmol(+)/kg), magnesium (10 cmol(+)/kg). The liquid limit was 560%, the plastic limit was 62% and the unit mass of the solid particles was 2.77 Mg/m^3 (Saba et al., 2014).

Cylindro-spherical pellets of bentonite were produced by compacting commercial bentonite powder in a mold of 7 mm in diameter and 7 mm in height. The initial suction (s = 135 ± 3 MPa) was measured in the laboratory with a chilled mirror dew point tensiometer (Decagon WP4C), corresponding to an initial water content w = 5% - 7%. The initial void ratio of the pellets was e = 0.306–0.386 (corresponding to a dry density of 2.00–2.12 Mg/m^3) and the degree of saturation was S_r = 55%–66%. The bentonite powder was produced by crushing pellets. An initial water content of 3% (smaller than that of the pellets) was found in the laboratory, corresponding to an initial suction s = 191 MPa (equally measured by a chilled mirror dew point tensiometer).

The saturation fluid was obtained by mixing the chemical components presented in Table 1 with distilled water until full dissolution. The resulting synthetic water had the same chemical composition as the pore water of the Callovo-Oxfordian claystone (a possible geological host rock for radioactive waste disposal in France) from the underground research laboratory in Bure (Wang et al., 2013a). The table shows a total mass of 5.677 g of salt per liter of solution, that corresponded to a salinity (mass of salt per mass of solution) lower than 1%.

2.2. Experimental set-up

A special set-up was designed to carry out X-ray CT observations on the pellet/powder mixture while wetting. The layout is presented in Fig. 1a. It consists of a transparent PMMA (PolyMethyl MethAcrylate) cell of 60 mm in inner diameter and 120 mm in height (corresponding to 1/10 scale of the SEALEX full-scale experiment). In the full-scale experiment, a horizontal borehole, with a 0.60-m diameter and 5.40-m length was excavated in the Tournemire URL, located in a Mesozoic sedimentary basin on the western edge of the French Causses. The clay-based core, which represented the engineered clay barrier, had a total length of 1.20 m. The laboratory specimen was installed between two porous stones and filter papers, placed at its top and bottom. The constant-volume condition for the pellet/powder bentonite mixture was ensured by the rigid PMMA cell and a top piston which could prevent axial swelling. The mixture was saturated from both sides (top and bottom), simulating the SEALEX in situ experiments (in the full-scale experiment, the clay-based core was saturated with water from both ends with a small pressure of 10 kPa). The axial swelling pressure was monitored during the test with a force transducer. The base and top were both equipped with a water inlet and an air outlet. Fig. 1b shows the PMMA cell positioned inside the X-ray microtomograph. The cell was connected to two reservoirs (located at the top and bottom of the sample) allowing water infiltration. The visualized parameter from X-ray CT observations was the linear attenuation coefficient, which was represented as a grey level. This parameter depended on density, the atomic number and the used X-ray energy.

2.3. Sample preparation and test protocol

The pellet/powder mixture was prepared by following the first protocol proposed in Molinero-Guerra et al. (2017) to obtain a homogeneous pellet/powder distribution within the sample at the target dry density (1.49 Mg/m^3). It consists in filling the cell by packets corresponding to one layer of pellets spread over its base and by adding the corresponding amount of powder (considering the proportion 80-pellet/20-powder in dry mass). The density of the mixture was checked with the mass of the bentonite (with known water content) used to fill the cell (with known volume). Moreover, some reference elements of 1500 PMMA spheres of 1.6 mm diameter (density 1.18–1.19 Mg/m^3) were randomly distributed within the sample during the preparation process. The purpose was to carry out quantitative analyses of bentonite powder displacement by DVC technique. DVC is a full-field measurement technique that basically consists in comparing two pictures at two different stages and calculating a displacement field. A detailed description of this technique can be found in Sutton et al. (2009) and Bornert et al. (2012).

The general experiment scheme consisted in scanning the pellet/powder mixture at different times during the saturation process. This resulted in a kind of incremental scans occurring during the hydration process: several X-ray CT observations were carried out at different times until an apparent homogeneous mixture was observed. Each scan took 19 h. Prior to each X-ray CT scan, the whole system shown in Fig. 1a was moved and installed inside the microtomograph.

The first step of the test consisted in opening the water inlet valves. Air inside the system was evacuated by opening the air outlet valve until no air bubble could be observed anymore in the outlets. Water was supplied through both the top and bottom of the sample, as in the SEALEX full-scale experiments, during the total time of the test with no pressure. Axial swelling pressure was recorded automatically by a force transducer and a numerical data logger.