Experimental characterizations of the hydro-mechanical behavior of a pellet/powder bentonite mixture

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ABSTRACT: Mixtures made up of bentonite and pellets are a possible candidate for sealing plugs used in deep radioactive waste disposal due to their low permeability, high swelling capacity, favorable properties with respect to radionuclide retention and operational advantages in terms of placement in situ (much easier than the pre-compacted bricks of bentonite/sand mixture). This paper presents an investigation on the hydro-mechanical behavior of a mixture of MX 80 bentonite pellets and powder (80/20 in dry mass) at different scales. An in-depth characterization of the mixture at its initial state revealed heterogeneity at both macroscopic and microscopic scales. An investigation of the water retention properties and of the microstructure of a single pellet of MX80 bentonite subjected to wetting/drying cycles under free swelling conditions was carried out by means of Mercury Intrusion Porosimetry and X-ray computed microtomography. A complete description of the microstructural distribution, the changes in water content, void ratio and degree of saturation of the pellet are presented. Hysteresis effects were also evidenced on the water retention curve.

1 INTRODUCTION

In the French concept of deep geological disposal for High Level and Intermediate Level Long Lived radioactive Wastes (HLW & ILLLW), the wastes are emplaced at great depths in large boreholes (HLW) and galleries (ILLLW) excavated in a low-permeability host-rock (Callovo-Oxfordian argillite, ANDRA, 2005). Sealing of these underground spaces to prevent potential pathways for water, gas and radionuclides migration is one of the key points for the long-term safety of the repository.

Sealing plugs made of powder/pellets bentonite mixture are considered as a possible option to close the galleries. Powder/pellets bentonite mixtures have low permeability, high swelling capacity and high radionuclide retardation properties. They are also easy to be transported and installed in the galleries and allow for reduced gaps between the host rock and the seal once hydrated. The high-density of both the pellets and the powder allows a high density of the seal and, as a result, an adequate swelling pressure. When installed in the gallery, powder/pellet bentonite mixtures are initially in an unsaturated state. They are afterwards hydrated under confined volume conditions due to the infiltration of pore water from the host rock, resulting in swelling that seals the gallery by filling the pre-existing voids.

In this context, and as part of the overall IRSN (French Institute for Radiological Protection and Nuclear Safety) R&D program that provides scientific background for its expertise on the disposal safety, the SEALEX (SEALing performance Experiments) project was launched to specifically focus on long-term performance of sealing systems. This project relies on a series of in situ experiments emplaced in IRSN´s Underground Research Laboratory (URL) at Tournemire located in a Mesozoic sedimentary basin on the western edge of the French Causses (South France) (Mokni & Barnichon 2016).

Various materials are being considered as seals in the SEALEX project. The present work focuses on a mixture made up of MX80 bentonite powder and pellets with a proportion of 20/80 in dry mass.

The microstructure of engineered barriers has been investigated by using mercury intrusion porosimetry (MIP), scanning electron microscopy (SEM, ESEM, Komine & Ogata 1999; Lloret et al. 2003; Montes 2007; Delage et al. 2006) and X-ray computed microtomography (μ-CT, Kozaki et al. 2001; Kawaragi et al. 2009; Van Geet et al. 2005).

The use of high-density bentonite pellets combined with powered bentonite has also been proposed to substitute the compacted bentonite blocks (Salo & Kukkola 1989; Dereeper et al. 2001). A characterisation of the fundamental properties of the MX80 bentonite pellets used in the Prototype Repository test is reported in Sugita et al. (2005). At a larger scale, a series of infiltration tests on a 50/50% FoCa bentonite pellet/powder mixture compacted at different dry densities were performed by Imbert and Villar (2006) to investigate the swelling capacity of the material. Hoffmann et al. (2007) described an experimental program performed to characterize the hydro-mechanical behaviour of compacted pellet mixtures packed at different dry densities used within the framework of the engineered barrier (EB) project (Rey et al. 2014). The pore size distribution of compacted pellet mixtures at different densities was obtained, with three main groups of pores identified, namely inter-aggregate, intra-aggregate and inter-pellets pores, respectively. The pore size density function of a single pellet was also obtained, with two pores population identified (intra-aggregate and inter-aggregate pores).

This paper deals with an experimental program aiming at studying the microstructural features of a pellet/powder mixture at various scales by using μ-CT observations coupled to MIP technique. An investigation of the water retention properties and of the microstructure of a pellet of bentonite submitted to wetting/drying cycles under free swelling conditions was carried out by means of Mercury Intrusion Porosimetry and X-ray microtomography. In particular, the role of suction in the internal structure of a pellet was investigated.

2 MATERIALS AND METHODS

2.1 *Material studied*

The investigated soil is a powder/pellet bentonite mixture with a proportion of 20/80 in dry mass. The bentonite comes from Wyoming, USA, with a high smectites content (80%) and some inclusions of non-clayey minerals. It was provided by the Laviosa-MPC company under the commercial name Expangel. Its cation exchange capacity (CEC) is 98 meq/100g. The major exchangeable cation is Na+, with a value of 52 meq/100g. The liquid limit is 560%, the plastic limit is 53% and the unit mass is 2.77 Mg/m^3 (Saba et al. 2014).

Pellets were produced by compacting the powder of MX80 bentonite in a mould of 7 mm diameter and 7 mm height (Figure 1). The fabrication water content is between 5% and 7% at a dry unit mass ρ_d = 1.998 Mg/m^3-2.12 Mg/m^3. The initial suction, s = 132.4 MPa, was measured in the laboratory with a chilled mirror dew point hygrometer (Decagon WP4) and the initial water content, w = 7.25%, was determined after drying the sample (pellet) at 105°C for 24h.

Figure 1. Pellet at initial state.

The MX80 bentonite powder used for the mixture was produced by crushing pellets of bentonite. It has to be noted that the initial water content after production is between 5% and 7%. However, a value of 3.17% was found after drying at 105°C during 24h. The initial suction, s = 190.9 MPa, was measured with the chilled mirror dew point hygrometer (Decagon WP4).

An in-depth characterization of bentonite pellets/powder at its initial state was carried out by Molinero Guerra et al. (2016). Heterogeneity was revealed in the internal structure of the pellet, in terms of density distribution of the clay minerals and other elements. At the macroscopic scale, a heterogeneous distribution of pellets and powder was identified.

2.2 *Experimental methods*

The determination of the water retention properties under free swelling conditions was carried out using the vapour equilibrium technique. The sample was periodically taken out of the desiccator for measuring suction and weight. Once the mass stabilised in the desiccator, the water content of the pellets was determined by oven drying at 105°C for a period of 24h. The specimen's volume was determined by hydrostatic weighing waxed specimens put in water-Wan et al. 2013). The void ratio, degree of saturation, dry density and volumetric deformation of the samples were calculated from their volume, mass and water content.

The microstructure of the pellet was investigated by determining the pore size distribution (PSD) curve. These curves were obtained on freeze-dried samples using an Autopore IV 9500 mercury intrusion porosimeter (Micromeritics) that operates at a maximum pressure of 230 MPa. Instantaneous freezing was carried out by plunging the pellet in slushy

nitrogen (-210°C) obtained by previously submitting it to vacuum (Delage 2007). In such conditions, there is no nitrogen boiling around the samples when plunging them into nitrogen, resulting in quick freezing and thus good microstructure preservation. In a standard fashion, the pore size distribution was interpreted assuming parallel, cylindrical nonintersecting pores of different radii.

In order to complete the investigation of the microstructure of a pellet of MX80 bentonite, several X-ray microtomography (µ-CT) observations were carried out. The X-ray sources parameters were 80kV and 40 µA. Voxel size was 4.41 µm. The samples were scanned using 1440 projections equally spread on 360°. After the reconstruction, 1298 horizontal slices were calculated (16 bit images; 1644 x 1292 pixels).

2.3 *Test programme*

A first series of tests was carried out to explore the wetting and drying paths starting from initial state (s = 135 ± 3 MPa, w = 7.25%, ρ_d = 2.06 ± 0.06 Mg/m^3) by submitting pellets to various suctions ranging from 0 to 262 MPa. The pellet wetted under 0 MPa (P9) did not reach the imposed suction value after 20 days, with a suction measured of 0.8 MPa. During a second series of tests, the pellets were first hydrated at a suction of 2.3 MPa (path I) prior to being dried under various suctions between 4.2 and 262 MPa.

The water content and dry density of the samples of both series of specimens when equilibrium was reached. MIP tests were performed on samples of the first series. In addition, X-ray computed microtomography observations were performed on specimens at the initial state and 9 MPa of suction. To follow the wetting path, ten specimens were hydrated through vapour phase by placing them in a desiccator containing distilled water. For the drying path, a value of 130 MPa suction was imposed to six pellets previously equilibrated at a suction of 2.3 MPa. During wetting/drying, mass and suction were measured.

3 EXPERIMENTAL RESULTS

3.1 *Water retention properties*

The water retention curve is shown in Figure 2. The hysteresis behavior can be observed with the drying path situated above the wetting path. The results are shown in Figure 3 in planes of i) degree of saturation versus water content (Figure 3a) and suction (Figure 3b) and ii) void ratio versus water content (Figure 3c) and suction (Figure 3d).

Figure 3b shows that, for the points of group A along the wetting path, the degree of saturation increased from 55.3% to 60% when decreasing suction from 135 MPa (initial state) to 82 MPa. At smaller suctions, the degree of saturation stabilizes at approximately 60%, because swelling (Figure 3e) compensates the increase in water content. Near saturation, an increase of degree of saturation is observed.

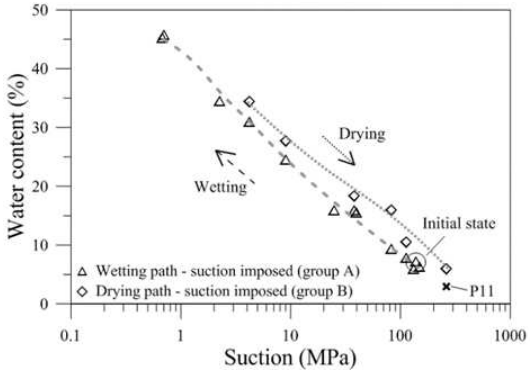

Figure 2. Water retention curve.

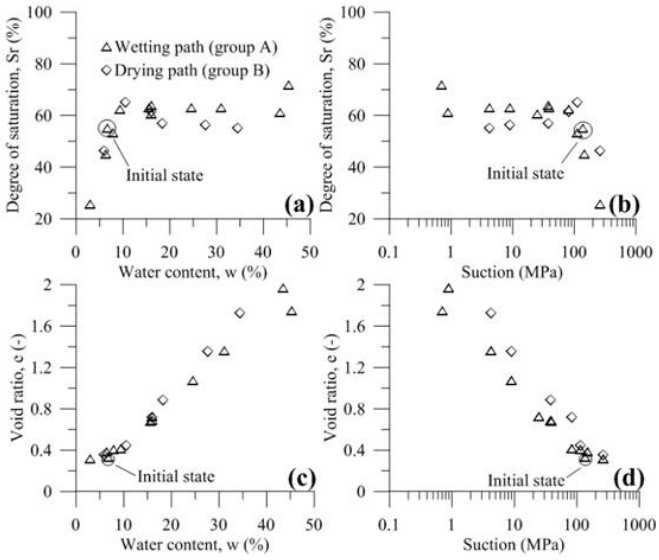

Figure 3. Volumetric behaviour obtained by suction-controlled method: (a) Degree of saturation versus water content, (b) Degree of saturation versus suction, (c) void ratio versus water content, (d) void ratio versus suction.

Hysteresis is observed when plotting the changes in void ratio with water content and suction (Figure 3c and d, respectively). For suctions higher than 82 MPa, the same void ratio is obtained for the wetting and drying paths. However, for suctions lower than 82 MPa, the drying path is situated above the wetting path. Along the drying path, starting from a wetted state near the saturation (s = 2.3 MPa), there is generation of some irreversible swelling strain.

3.1 *Microstructure*

The results of MIP tests on pellets at the initial state (135 MPa of suction), dried at 149 MPa and 262 MPa of suction are presented in

Figure **4**.

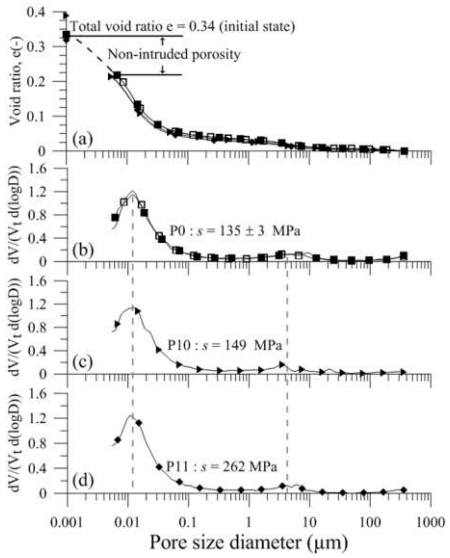

Figure 4. MIP tests results for pellets at the initial state, 149 MPa and 262 MPa of suction.

4 Two pellets at initial state were tested in order to investigate the repeatability of the method adopted and possible heterogeneity between pellets. It appears from

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7 FIGURE **4** that the results obtained for both pellets are similar. In all cases, a difference between the maximum void ratio intruded by mercury and the total void ratio of the pellets is observed in the cumulative curves (

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FIGURE **4**a). In the case of heavily compacted smectite bearing materials, the porosity not intruded at the highest mercury pressure, called here infra-porosity, is related to small pores with entrance diameter smaller than 4 nm (Delage et al., 2006). The comparison of the curves shows that the various high suctions imposed here (135, 149 and 262 MPa) have no significant effect on the pore size distribution of the pellets, that are defined by a pore population with an average entrance diameter of 11.9 nm for samples at 135 MPa and 149 MPa and 10.7 nm for the pellet at 262 MPa. Cumulative and density functions curves also show a pore population at diameters around 4 – 5 μm.

Figure 5 presents the pore size distribution curves of pellets hydrated from initial state (135.5 MPa) to suctions higher or equal than 25 MPa along the wetting path: 113 MPa, 82 MPa, 40 MPa, 38 MPa and 25 MPa. The curves show that the average entrance pore radius stays equal to 10.7 nm at 113 MPa, whereas it increases at values between 14.9 and 16.6 nm for suctions equal or smaller than 82 MPa. Also, the pore population around 4 – 5 μm, previously observed at large suctions, is still observed at suctions of 113 and 82 MPa.

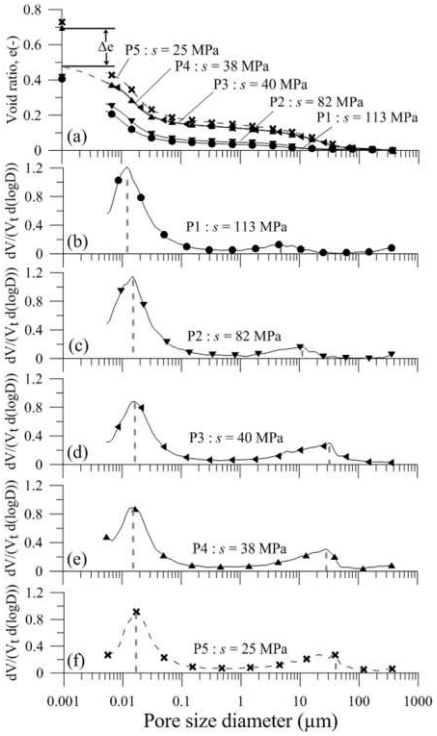

Figure 5. MIP tests results for pellets hydrated at 113 MPa, 82 MPa, 40 MPa, 38 MPa and 25 MPa of suction.

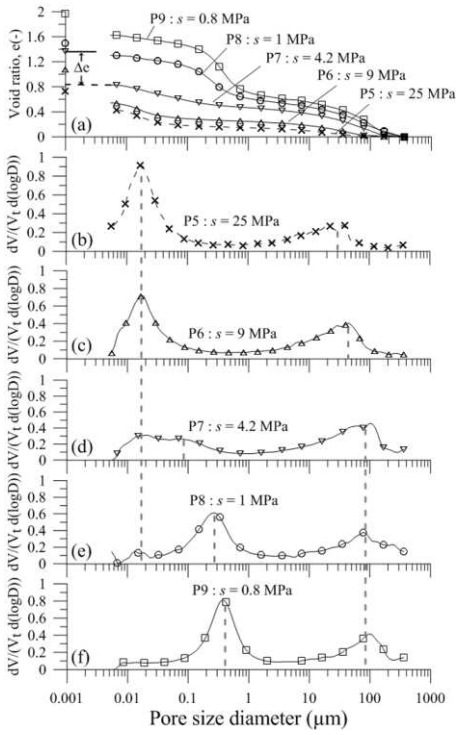

Figure 6. MIP tests results for pellets hydrated at 25 MPa, 9 MPa, 4.2 MPa, 1 MPa and 0.8 MPa of suction.

Figure 6 presents the pore size distribution curves of pellets submitted to suctions equal or lower than 25 MPa along the wetting path. Whereas cumulative curves at 25 and 9 MPa remain comparable to that of the former series, the curve at 4.2 MPa looks like a transition towards the curves at lower suction, with a decrease in the peak previously observed at 19 nm and the development of pores with a diameter up to 400 nm. At very low suction (1 and 0.8 MPa), no significant porosity is observed below 0.1 µm and the significant swelling is due to the development of pores with average diameters of 290 nm (suction 1 MPa) and 400 µm (suction 0.8 MPa).

Figure 7 presents two horizontal sections of a pellet at its initial state (135 MPa of suction) (sections I, II) and two other horizontal sections (III, IV) of a pellet hydrated at 9 MPa of suction. A 3D reconstruction for both cases as well as the fissure network for a region of interest are presented in Figure 8: a total view as well as a vertical section for both cases are presented. A clear difference is observed for a pellet at its initial state and after hydration: for the first case, some cracks are observed at the edge to the pellet, probably due to the swelling of the specimen during the storage in the laboratory combined with the fabrication process. After hydration, cracks are observed everywhere within the pellet. As mentioned above, these cracks within a pellet are induced by significant swelling under free swelling conditions.

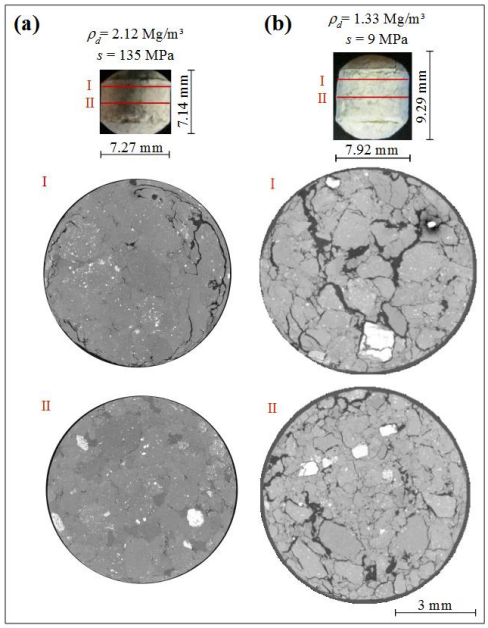

Figure 7. X-ray computed microtomography (µ-CT) observations – horizontal section: (a) pellet at its initial state; (b) pellet hydrated at 9 MPa of suction.

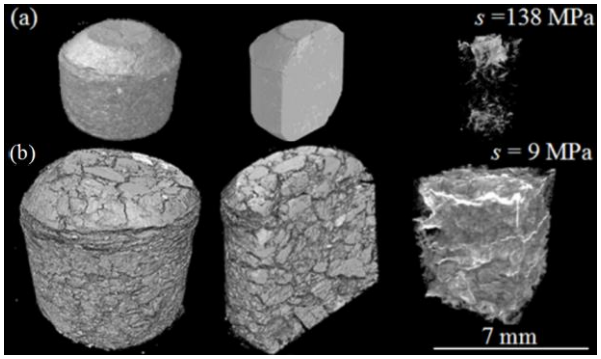

Figure 8. X-ray computed microtomography (µ-CT) observations – 3D reconstruction image and network of fissures.

10 DISCUSSION

Based on µ-CT observations combined with pore size distribution curves at different suctions, it has been demonstrated that swelling is not homogeneous among the clay matrix but corresponds to the development of cracks. While wetting, the pellet is taking water and swelling at the same time, which results in a constant value of degree of saturation under 82 MPa suction (Figure 3a, b). During this phase, cracks are developed. Near saturation, cracks are filled with water and an increase of degree of saturation is observed.

The changes in PSD curves in the small pore range are governed by different phenomena at a different scale, with average pore diameters of 11 nm at initial state and also up to 262 MPa suctions. Indeed, the application of higher suctions along the drying path has no effect on this pore population, like also the decrease from 135 to 113 MPa suction. The intruded porosity is the same at suctions between 262 and113 MPa, and it slightly increases at 82 MPa.

From the investigation of the microstructure, we can conclude that hydration is governed by the progressive placement of layers of water molecules along the surface of the elementary clay layer inside the clay particles, with one layer in dry conditions (initial state of the pellet) and ending up with a maximum number of four layers. The swelling mechanism starting from high suction is the results of two combined phenomena: (i) the progressive insertion of successive layers of water molecules in the interlayer spaces inside the particles and (ii) the subdivision of particles into thinner ones that are made up of a smaller number of stacked layers. This two phenomena combined give rise to an inter-particles porosity that develops inside the aggregates, which explains the shifting observed for values of suction under 4-10 MPa. This value of suction can be considered as a threshold, because a new shape of the pore size distribution curve is observed at this state (specimen P7). Note that Delage et al. (2006) found a threshold value of 7 MPa of suction for a compacted bentonite. The increase of the mean size diameter of big pores during hydration corresponds to the fissures observed while wetting under free swelling conditions (Figure 7 andFigure **8**).

11 CONCLUSION

Experimental data from different testing methodologies on a pellet of MX80 bentonite allowed the analysis of the effects of wetting/drying cycles under free volume changes on fabric evolution and water retention properties. The main outcomes of the study are summarized as follows.

The significant difference observed between the main drying path and the main wetting path confirmed the hysteresis of the water retention properties of the MX80 bentonite. The observation on the degree of saturation versus suction showed a constant value of degree of saturation under a certain suction. This result can be explained by the fact that the pellet is swelling and keeping water at the same time. Near saturation, cracks are filled with water and an increase of degree of saturation is observed.

- When wetting at suctions lower than 10 MPa, a change of the shape of PSD curve is observed, and under such low suctions, near saturation state is reached, leading to a shifting of the mean intra-aggregate pore size diameter.

Molinero Guerra et al. (2016) concluded that the mixture composed of powder/pellets of bentonite is characterized by a heterogeneous structure at the initial state, with the presence of voids between the pellets. Results presented in this work allow to understand the behaviour of pellets located in the mixture while wetting, as they start swelling under pseudo-free swelling conditions.

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