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Abstract

Compacted bentonite-based materials have been proposed as possible sealing and backfill materials in geological repositories for the high-level radioactive waste disposal in several countries. During the long time lifespan of a repository, as the chemical composition of porewater can change, the swelling and sealing capacity of the material may also change. From a point of view of storage safety assessment, it is important to evaluate this possible change of swelling capacity. In this study, a mixture of bentonite and crushed Callovo-Oxfordian claystone was investigated. The long-term effect of pore water chemistry on the swelling pressure was studied at constant-volume conditions for 700 days. Distilled water and synthetic water having the chemical composition similar to that of in-situ pore water were used for hydration. The results obtained in an initial period of 100 hours revealed no significant influence of the water composition on the swelling pressure evolution, and the maximum swelling pressures observed were close to 4.30 MPa for a dry density of 1.70 Mg/m^3 . Over a longer time period, on the contrary, the swelling pressure eventually decreased for all samples, especially for the sample saturated with synthetic water. In addition, comparison of a one-step soaking test with a multi-step soaking test showed no wetting procedure effect on the long term swelling behaviour. All the results are analysed by considering the physico-chemical interaction between the minerals of claystone, minerals of bentonite and different fluids involved.

Keywords: Radioactive waste disposal; bentonite based materials; pore water chemistry; time dependence; swelling pressure

1 INTRODUCTION

Compacted bentonite-based materials have been proposed as possible sealing/backfill materials in deep geological repositories for high-level radioactive wastes (HLW) in several countries. Due to their favourable swelling characteristics, these materials are expected to fill up all voids left in the system after construction and emplacement of the waste packages. An important long-term safety function of bentonite-based barriers is to ensure a relatively impermeable zone around the high-level radioactive waste thereby limiting groundwater flow and waste package degradation rates and, ultimately, waste leaching rates. This role termed as “sealing” (Komine, 2004; Komine and Ogata, 2003) is essential to ensure the overall safety of the geological disposal: the disposed waste is isolated from the biosphere over long time. For this purpose, the swelling property becomes a key factor in the design and fabrication of the sealing/backfill materials, and needs to be studied in depth.

The swelling properties (i.e. swelling pressure or swelling potential) of bentonite-based materials have been widely investigated (Pusch, 1982; Komine and Ogata, 1994, 2003, 2004a; Delage et al., 1998; Agus and Schanz, 2005; Komine et al., 2009). They depend strongly on the initial state such as dry density and water content (Komine and Ogata, 1994; Villar and Lloret, 2008). After being installed in the field, especially in some geological host formations relatively rich in salts (e.g. salt formation, claystone, granite formations, etc.), site water of certain salinity can interact with bentonite (Herbert et al., 2008); as a result, the hydro-mechanical behaviour of the bentonite-based materials and in turn the performance of the barrier may be notably affected. Moreover, the water composition can change over time.

Frequently, bentonite/sand mixtures are considered for the reason of good control of swelling pressure, enhanced thermal conductivity as compared to pure bentonite and a better mechanical resistance. For instance, a 70/30 bentonite-sand mixture was proposed as buffer

material in Japan while a 50/50 bentonite/sand mixture was suggested in the Canadian repository concept (Dixon et al., 1985). In France, Callovo-Oxfordian (COx) claystone has been considered as a possible geological host-rock for high-level radioactive waste disposal, and the mixture of bentonite and crushed Callovo-Oxfordian (COx) claystone is proposed as a possible sealing/backfill material for environmental and economic reasons (Andra, 2005; Tang et al., 2011a; Tang et al., 2011b; Wang et al., 2012). If physico-chemical interactions essentially take place only between groundwater and bentonite in the case of sand/bentonite mixture, it is not the case for the bentonite/claystone because of the additional interactions between these two materials and the water. Due to the low permeability of the bentonite-based materials, these interactions might develop over long time and the resulting effects will not be immediate. Hence, particular attention should be paid to their long-term behaviour.

This study focuses on the swelling pressure of the compacted mixture of MX80 bentonite and crushed COx claystone. The long term effect of water chemistry on the swelling pressure and microstructure were investigated after 700 days in contact with water. Emphasis was put on the physico-chemical interaction between the minerals of claystone, minerals of bentonite and different fluids involved. The wetting procedure effect on the long term swelling behaviour was also investigated by comparing a one-step soaking test with multi-step soaking test.

2 MATERIALS AND METHODS

2.1 Materials

The bentonite used in this study is a commercial MX80 Na-bentonite, which contains large quantities of montmorillonite (75-90%). Silt is the dominant remaining fraction, which mainly consists of quartz and feldspars as well as micas, sulphides, and oxides (Pusch 1982). Table 1 details the main mineralogical components of MX80 bentonite obtained by several authors.

The total cation exchange capacity (CEC) is between 0.78 and 0.85 meq/g (Table 2), the cation population at exchange positions is mainly composed of Na^+ (60.0–67.0 meq/100g), other population being much lower: Ca^{2+} (5.0–8.0 meq/100g), Mg^{2+} (3.0–4.0 meq/100g) and K^+ (0.2–1.3 meq/100g). The grain size distribution (Fig. 1) determined by hydrometer (AFNOR NF P94-057) shows that the fraction of clay-size particles ($< 2 \mu\text{m}$) is 84%. The bentonite tested has an average specific gravity of 2.76, a liquid limit of 520%, and a plastic limit of 42%.

Callovo-Oxfordian (COx) claystone was taken at 490-m depth from the Underground Research Laboratory (URL) at Bure in eastern France. It contains 40–45% clay minerals (mainly interstratified minerals of illite–smectite), 20–30% carbonates (mainly calcite) and 20–30% quartz and feldspar (Hoteit et al., 2000; Lebon and Ghoreychi, 2000; Zhang et al., 2004). The in-situ water content is 2.8–8.7 %; the bulk density is 2.32–2.61 Mg/m^3 and the specific gravity is 2.70 (Hoteit et al., 2000; Tanget al., 2011a). The excavated claystone was air-dried and crushed into a powder. Fig. 1 depicts the grain size distribution of the crushed powder obtained by dry sieving; it contains 18% of fine grains (0.08 mm). The grain size distribution determined by hydrometer confirms that the content of clay-size particles ($< 2 \mu\text{m}$) is 40%.

Both distilled water and synthetic water were used for hydration. The chemical composition of the synthetic water (see Table 3) is similar to that of the groundwater at Bure site. The pH values are between 7.00 and 7.28; the concentration of Na^+ is twice that of Ca^{2+} and Mg^{2+} and four times that of K^+ (Gaucher et al., 2006; Marty et al., 2010).

2.2 Sample preparation

In this study, all tests were performed on samples of compacted bentonite/claystone mixture with a bentonite content of 70 % in dry mass. Bentonite and claystone powders, with the

initial water contents of 11.8 % and 2.64 % respectively, were first carefully mixed prior to compaction. Samples were statically compacted to a dry density of 1.70 Mg/m^3 in a metallic cell (70 mm in internal diameter) at a controlled rate of 0.05 mm/min. After compaction, the sample height is about 10 mm. The total suction measured in these specimens using a hygrometer was $s = 90 \text{ MPa}$. The specimens are then carefully introduced into constant-volume cells (having the same diameter as the compaction cell) for the swelling pressure experiment.

2.3 Experimental methods and programme

The constant-volume cell used in this study is presented in Fig.2. It consists of three parts: (1) the bottom part containing a porous stone and a drainage system; (2) the middle cell (70 mm inner diameter, 10 mm height) used to prevent radial swelling, with two air outlets; (3) the top part incorporating a total pressure sensor (working pressure of 5 MPa) to monitor the swelling pressure. The sample was wetted by connecting the water inlet to a water reservoir or a suction control system using vapour equilibrium technique at an ambient temperature of 20 ± 1 °C. More details can be found in Tang et al. (2011b), Wan et al. (2012).

Four swelling pressure tests were performed using the constant-volume cell (Table 4). In tests LT01 and LT02 (LT stands for long term), the samples were saturated with synthetic water and distilled water, respectively. The duration of these tests carried out to study the long-term effect of water chemistry on the swelling pressure was 700 days. In test LT03, three decreasing suctions (57 MPa, 38 MPa and 12.6 MPa) were first applied in steps using vapour equilibrium technique, prior to distilled water flooding in the last stage. This test also lasted 700 days. This test was performed to investigate the effect of the wetting procedure on the long term swelling behaviour by comparison with test LT02. Test ST (ST for short term) was performed by directly injecting synthetic water as in test LT01, but with a shorter duration of 100 hours.

After certain hydration time (see Table 4), the specimen was taken out of the cell for the analysis of pore-size distribution by Mercury Intrusion Porosimetry (MIP).

3 EXPERIMENTAL RESULTS

Fig.3 presents the results from tests LT01, LT02, and ST during the first 100 hours after the injection of water. The three curves are very similar, showing negligible effect of water chemistry for this duration and also the good repeatability of the tests. With water infiltration, swelling pressure first increased very quickly; after about 20 hours the swelling pressure reached a first plateau at about 3.4 MPa. After 32 hours, the swelling pressure started to increase again and reached a second plateau after 100 hours. The final values were in the range of 4.30 – 4.37 MPa for the three tests.

The results of test LT03 are presented in Fig.4. The application of the first suction of 57 MPa resulted in a swelling pressure of 0.57 MPa. Then, the second suction of 38 MPa was applied and the swelling pressure reached 1.43 MPa. With the third suction of 12.6 MPa the swelling pressure increased to 2.61 MPa. The zero suction applied by direct contact with distilled water led the sample to a maximum value of 4.39 MPa swelling pressure. This value is quite close to that from test LT02 (4.37 MPa) in which the sample was directly put in contact with distilled water. This indicates that the wetting procedure did not affect the short-term swelling behaviour for this material.

In Fig.5, all results from the three tests LT01, LT02 and LT03 during 700 days are presented. It appears that over this long period, the swelling pressure decreased somewhat for all samples, especially for the sample saturated with synthetic water (LT01). The final swelling pressure was 3.95 MPa (corresponding to a decrease of 9%), and 4.19 MPa (decrease of 3%) for

167 samples saturated with synthetic water (LT01) and distilled water(LT02 and LT03),
168 respectively.

169 The pore size distribution curve for all the samples taken at the end of the tests are presented
170 in Fig. 6. It is observed in Fig. 6a that the final values of intruded mercury void ratio ($e_m =$
171 intruded mercury volume over soil solid volume) are between 0.4 and 0.5, much lower than
172 the global void ratio ($e = 0.61$). This shows that a large amount of porosity is inaccessible to
173 mercury. Comparison between the curves obtained after different durations clearly shows that
174 the amount of accessible porosity after 700 days is larger in both cases of distilled water
175 (LT02 and LT03) and synthetic water (LT01) than after 100 hours with synthetic water. In
176 addition, for samples hydrated for 700 days, more quantity of accessible porosity is observed
177 in the case of synthetic water (LT01).

178 The incremental pore volume $de_m/d\log(d)$ (Fig. 6b) refers to the volume of mercury that
179 intrudes into the pores between pressure increments. For all samples a typical bimodal
180 porosity can be observed; thus, two pore groups can be defined: a group of micro-pores
181 having a mean size of $0.02\ \mu\text{m}$ and a group of macro-pores having a mean size of $20\ \mu\text{m}$.
182 After 700-day saturation (LT01, LT02 and LT03), the macro-pores and micro-pores quantity
183 increased when comparing to the short-term test (ST). Change in macro-pores is more
184 significant especially for the samples saturated with synthetic water (LT01). For tests LT02
185 and LT03 which were hydrated with distilled water, very similar curves were observed
186 suggesting no effect of the wetting procedure on the pore-size distributions. As far as changes
187 in pores size are concerned, it can be observed that over time the group of micro-pores had a
188 size slightly decreased. On the contrary, the group of macro-pores had a size slightly increased.
189 Changes for test LT01 were more marked.

4 INTERPRETATION AND DISCUSSION

Various studies showed that the swelling capacity of bentonite is lower in saline water; the higher the dissolved salt concentration, the lower the swelling pressure. This phenomenon becomes less pronounced in case of high dry density (Studds et al., 1998; Karland et al., 2005; Suzuki et al., 2005; Karland et al., 2006; Castellanos et al., 2008; Katsumi et al., 2008; Siddique et al., 2011). In general, with a high void ratio (low dry density) and/or high salt concentrations, the influence of pore water chemistry on bentonite behaviour is significant and can be qualitatively explained by the diffuse double layer (DDL) theory (Karland, 1997; Mata, 2003; Castellanos et al., 2008). For dense bentonite, since there is little water present between adjacent clay platelets, there can be little or no development of a DDL associated with the clay particles (Dixon, 2000; Pusch and Yong, 2006); thus it becomes difficult even impossible to describe the bentonite behaviour by the DDL theory.

In this regard, Karland et al. (2005) studied the effect of salinity on the swelling pressure of MX-80 bentonite, and noted that the effect of salinity of the saturating fluid was relatively lower with a higher density. For MX80 bentonite, Dixon (2000) also concluded that at an effective bentonite dry density (bentonite dry density in the bentonite/sand mixture) higher than 1.22 Mg/m^3 , changes in water chemistry do not significantly affect the swelling behaviour due to the small number of water molecule layers between clay sheets. Confirmation was made by Castellanos et al. (2008) on the FEBEX bentonite: an increase in salt concentration reduced the swelling pressure, but this change was much less marked for high density. It can be deduced that for the studied bentonite/claystone mixture, the high density (1.70 Mg/m^3) and the low salinity of the synthetic water (see Table 3) representative

of the present in situ conditions resulted together in the negligible effect of water chemistry on the swelling pressure in short term.

Basically, the swelling pressure of bentonite is mainly related to the crystalline swelling and double layer swelling. Crystalline swelling is caused by the hydration of exchangeable cations (K^+ , Na^+ , Ca^{2+} , Mg^{2+}) between mineral layers that have a structure with one alumina octahedral sheet sandwiched between two silica tetrahedral sheets. It is a process which involves the adsorption of a maximum number of hydrates depending on the nature of cations. After three to four water monolayers, i.e. at interlayer separation distances ≥ 1 nm, the surface hydration becomes less significant and the electrical double-layer repulsion becomes the main swelling mechanism (Bradbury and Baeyens, 2003; Suzuki et al., 2005). For clay at high density, the low amount of water taken up is to all intents and purposes pseudo-crystalline interlayer water, and insufficient to form the DDL (Pusch and Yong, 2006). Therefore, the swelling pressure is mainly governed by the crystalline swelling (interaction between the layer surfaces and water) and the double layer repulsion makes only a limited contribution. In this case, the exchangeable cations are the key elements that control the clay-water interaction (Abdullah et al., 1997).

As far as the pore water is concerned, it may significantly affect the chemical compositions of clays through the exchangeable cations (Yukselen et al., 2008). The cation exchange reaction is mainly controlled by the exchange capacity of the clay minerals (Mata, 2003); the ease with which a cation of one type can replace a cation of another type depends mainly on the valence, relative abundance of the different cation types, and the cation size. Other factors being equal, the higher the valence of the cation, the higher the replacing power; for cations of the same valence, the replacing power increases with the size of the cation (Laine and Karttunen, 2010). Atypical replaceability order is: $Na^+ < K^+ < Mg^{2+} < Ca^{2+}$ (Mitchell, 1976; Pusch, 2001; Mata et al.,

2003). According to this order, for the Na-bentonite as in this study, it tends to change from sodium (Na^+) to other types (i.e. K^+ , Mg^{2+} or Ca^{2+}) depending on the cations present in the permeating water.

One of the most common cation exchange reactions in the Na-bentonite is the one involving sodium and calcium (Mata, 2003). Mata (2003) investigated the evolution of Ca^{2+} and Na^+ in a specimen of sodium MX80 bentonite hydrated with saline water (NaCl and CaCl_2 , 50/50 by mass) and distilled water. Analysis of the collected pore fluid showed an increase of sodium concentration and a decrease of calcium concentration as compared to the injected water. This suggests that the sodium bentonite was transformed to a calcium bentonite. Montes-H. et al. (2004, 2005) investigated the chemical transformation of Na/Ca-MX80 bentonite ($\text{Na}_{0.18}\text{Ca}_{0.10}$) in a representative pore water ($\text{pH} = 7.3$) from Bure site (-180 m), the Na/Ca- to Ca-montmorillonite conversion was identified as the main chemical transformation. At a constant temperature, this transformation is affected by the groundwater composition (pH and cation concentrations) and solid/liquid ratio as well as the reaction time (Mitchell, 1976; Herbert et al., 2004; Fernández and Villar, 2010; Laine and Karttunen, 2010). For the short term swelling pressure test in this study, it was actually the low cation concentration (low salinity of synthetic water mentioned above) and high solid/liquid ratio (i.e. high density) that led to the negligible effect of water chemistry on the swelling pressure. Nevertheless, after a long time period of reactions, the effect became evident because the presence of Ca^{2+} in Bure site water (see Table 3) enabled part of Na-montmorillonite to change to Ca-montmorillonite which has a much lower expansibility. This process resulted in a decrease of swelling pressure over time as shown in Fig. 5.

The transformation from sodium to calcium montmorillonite is critically dependent on the amount of available calcium (Ca^{2+}) in the pore water (Muurinen and Lehtikoinen, 1999;

Fernández et al., 2004, Fernández and Villar, 2010). For the sample saturated with distilled water, the pore water chemistry evolved over time to reach equilibrium with the claystone minerals (the claystone contains 20–30% carbonates, mainly calcite). Consequently, it was as if diluted Bure site water infiltrated to the MX80 bentonite, leading to the degradation of swelling pressure. However, with a much lower cation concentration (only 30% claystone in the mixture), the decrease of swelling pressure was less significant than in the case with synthetic water.

For the sample first wetted by three steps of suction (LT03), after the sample was flooded with distilled water, the maximum swelling pressure was found close to that obtained by water-flooding the sample directly (LT02), indicating a negligible effect of wetting procedure on the swelling pressure (Figs. 3 and 4). This phenomenon can be explained by the mechanism identified by Cui et al. (2002): when hydrating by decreasing suction (57–38–12.6 MPa) under confined conditions, the macro-pores were progressively clogged by the invasion of exfoliated clay particles, whereas the micro-pores remained almost unaffected. The micro-pores started to change only when the water saturation is approached. This suggests that step-wetting by suction control and direct flooding with water may lead to similar microstructures after saturation, thus similar maximum swelling pressures. After 700-day hydration with distilled water, not only similar swelling pressures (Fig. 5), but also similar pore-size distributions were observed for tests L02 and L03 (Fig. 6), which enhanced this conclusion. In addition, the repeatability of swelling pressure test and the MIP test was verified.

It was noted that the chemical effect on the HM behaviour of clayey soils is due to changes at different structural levels and the interaction between them (Mata 2003). From the pore-size distribution observation in this study, it can be seen that the amount of accessible porosity increased after a long time period for both distilled (LT02 and LT03) and synthetic water

(LT01), with more increase for synthetic water (Fig. 6). If we relate these phenomena to the swelling pressure, it is clear that the sample with lower swelling pressure had a higher intruded mercury void ratio in both micro- and macro-pores. For samples with higher swelling capacity, more interlayer hydration occurred, leading to a constriction of accessible pores. When referring to the incremental pore volume, the same tendency can be identified, for the sample with higher swelling pressure, both the macro-pores and micro-pores decreased. However, most of changes in microstructure occurred in macro-pores family. The same observation can be made in terms of pore size changes. This is in agreement with the observation made by Mata (2003) who conducted MIP test on the compacted sodium bentonite MX80 and sand mixture (70/30) (dry density of 1.37 and 1.67 Mg/m³) saturated with distilled water and saline water, and found that the saline water effects were significant on the macro-pores, the micro-pores remaining almost unaffected.

5 CONCLUSION

Long term swelling pressure tests and MIP tests on a bentonite/claystone mixture were performed. The effects of water chemistry as well as the hydration procedure on the swelling pressure were investigated in both short and long terms. The results are analysed by considering the physico-chemical interaction between the minerals of claystone, the minerals of bentonite and the different pore water chemistries involved.

There was no obvious effect of water chemistry on the swelling pressure in short term (100 hours) due to the high dry density of the bentonite-based material and the low salinity of the synthetic water. However, after a long period of 700 days, the swelling pressure decreased for all samples, especially for the sample saturated with synthetic water.

Different hydration procedures led to comparable long term swelling behaviour and similar pore-size distributions.

The effect of the chemical composition of the pore water on the swelling pressure can be interpreted in terms of changes in microstructure. The sample with higher swelling pressure showed less macro and micro-pores associated with a higher interlayer hydration.

From a practical point of view, the specification and the design of the sealing/backfill material are often made based on the swelling properties measured in short term. However, a non-negligible decrease (9%) of swelling pressure was observed after 700 days even with a water of low salinity. This expected reduction of swelling pressure has thus to be taken into account when designing a repository to ensure that the bentonite-based barrier will also meet the requirements on which its long term performance rests. Considering the possible pore water chemical composition changes over the very long term lifespan of the repository is also recommended to avoid deterioration of the swelling capacity of these materials and thus of the self-sealing capacity of the system.

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