Hydro-chemo-mechanical behavior of a sand/bentonite mixture upon wetting paths

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ABSTRACT: Sand/bentonite (S/B) mixtures are considered as backfilling materials for nuclear waste storage. The paper presents an experimental campaign that was carried out to investigate the swelling behavior of a S/B mixture, under different hydro-chemo-mechanical loadings. Performed experiments include free and confined swelling tests and controlled-suction confined swelling tests. Different dry densities and waters of different salinity were considered in the experiments. The fabric evolutions associated with the swelling behaviour was investigated by combining mercury intrusion porosimetry analyses and scanning electron microscope photomicrographs. The swelling potential was found to be sensitive to variations in compaction dry density, applied matric suction and pore water salinity. The pore size distribution (PSD) of the compacted specimens was found to be bimodal, irrespective of the dry density or of the water used for the preparation of the mixture. Tests on specimens compacted at the same dry density and saturated with different types of water under constant volume conditions have revealed how with increasing salinity of the solution, the final PSD moves from a mono-modal to a bi-modal distribution. The obtained data combined with results from other studies allowed to define a trend for the calculation of the swelling pressure as a function of the relative dry density of the bentonite in the mixture.

1 INTRODUCTION

Engineered barriers are fundamental components for the geological disposal of nuclear wastes. In the Swiss concept for low and intermediate level wastes, sand/bentonite (S/B) mixtures at a low bentonite content (20–30% of bentonite) have been selected as possible backfilling materials for waste isolation (Nagra, 2008).

Bentonite-based materials are selected as buffer and backfilling materials because of the presence of a large amount of smectite minerals. The high activity of smectite guarantees its capacity to adsorb water and hold it tightly both in its interlayer pores and on its external surface. The resulting swelling behavior is convenient in order to seal technological gaps in the waste repository and to limit preferential flow paths. However, the swelling capacity of the smectite at the particle level and the related swelling capacity at the macroscopic level, are affected by the environmental conditions in which the bentonite is emplaced. In particular, a reduction of the swelling capacity is expected with increasing salinity of the pore water (e.g. Pusch, 2001; Castellanos et al., 2008; Komine et al., 2009).

In this context, this paper presents selected results from a broad experimental campaign performed with the objective of understanding and quantifying the effects of the microscopic features of bentonite-based materials on its macroscopic swelling potential when subjected to different Hydro-Chemo-Mechanical (HCM) loadings. Free, confined and controlled-suction swelling tests were performed using different salinities of the pore fluid and initial dry densities. The microstructure of the S/B mixture was studied in saturated and unsaturated conditions and under different chemical and mechanical conditions by mercury intrusion porosimetry (MIP) and by high-resolution scanning electron microscopy (SEM). Further details on the experimental campaign can be found in Manca et al. (2016).

2 MATERIALS AND METHODS

2.1 *The Sand/Bentonite mixture*

A mixture of quartz sand and MX-80 bentonite was used, with a proportion of 80/20 in dry mass. The index properties for the mixture are: density of the solid particles 2.67 Mg/m^3; liquid and plastic limits 420% and 65%, respectively. Figure 1 shows SEM photomicrographs of the constituents of the mixture. The bentonite has a smectite content of 85%, hygroscopic water content of 5–6% at a temperature of 21°C and a relative humidity of 35–45%, and was prepared in granular form. The apparent grain size distributions of the MX-80 granular bentonite, the sand and the mixture are reported in Figure 2.

The dry sand and the granular bentonite were sieved at 0.5 mm and then mixed manually. Water was gently added with a spray gun with continuous mixing to reach a water content of 11%, which corresponds to the optimum water content for a dry density of 1.86 Mg/m^3, for a dynamic compaction with an energy of 3.4 J/cm^3. Finally, the mixture was stored in hermetic containers for at least 3 days to ensure moisture equalisation.

2.2 *Preparation of the specimens*

Specimens for the hydro-mechanical tests were prepared by static compaction of the mixture at a constant rate of 0.5 mm/min. Specimens for microstructural analyses were compacted to two target dry densities inside an isochoric cell designed in house (Seiphoori et al., 2014); the wetting to the desired water content was performed by injecting water with a syringe; after wetting, the cell was hermetically wrapped in paraffin for 15 days to allow the internal moisture equalisation; cylindrical specimens of 8 mm diameter and 7 mm high were re-cored using a sharp steel tube immediately before the freeze-drying procedure, to limit disturbance and desiccation of the specimens.

Four different waters were used to prepare the mixture and for the subsequent wetting of the specimens.

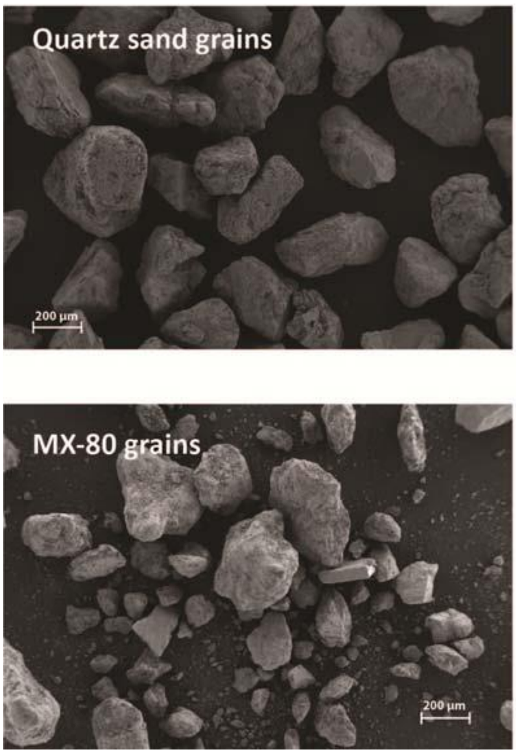


Figure 1. SEM photomicrographs of the quartz and the MX-80 bentonite grains.

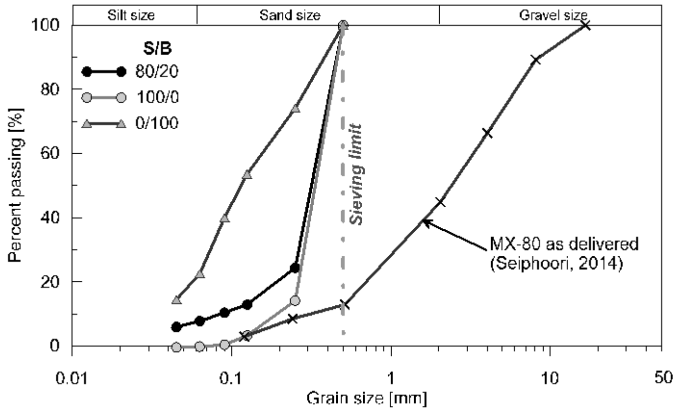


Figure 2. Apparent grains size distribution of the tested material and its two solid components. The bentonite was prepared in granular form.

Distilled water (DW) was used to allow the development of the maximum swelling of the bentonite. A synthetic water (SW) reach in sodium chloride (about 165 mmol/Kg of H_2O), having the same composition of the in situ pore water that can be found in Opalinus Clay formation, was used to reproduce the real conditions of a repository (details in Traber, 2011). The two other fluids were sodium chloride solutions prepared at 1 mol/l (1 M NaCl) and 4 mol/l (4 M NaCl) concentrations. The osmotic suction of the three salty waters were 0.9 MPa for the SW (Ferrari et al. 2014a), 4.9 MPa for the 1 M NaCl solution, 22.6 MPa for the 4 M NaCl solution (Witteveen et al. 2013).

2.3 *Swelling tests*

Conventional free and confined swelling tests, and controlled-suction confined swelling tests were performed on the S/B mixture, compacted to target dry densities in the range of 1.3 – 1.9 Mg/m^3 and wetted with the four types of water described before. To ensure constant volume conditions in the constrained swelling tests, the cell was positioned inside a high-rigidity frame.

The controlled suction swelling tests were performed in a cell allowing the control of matric suction through the axis translation technique, maintaining a constant air pressure and varying the pore water pressure according to the target matric suction value. A high-air-entry ceramic disc (air entry value (a.e.v.) 500 kPa) was mounted at the bottom of the cell. Each matric suction step was maintained until suction equilibrium conditions within the specimen were achieved (Airò Farulla & Ferrari, 2005). Water volume exchanges through the specimen were measured during the equalisation steps. Similarly to the conventional swelling test, the cell is placed inside a high-rigidity frame to maintain isochoric conditions.

Swelling pressure was measured with a load cell placed at the top of the specimen.

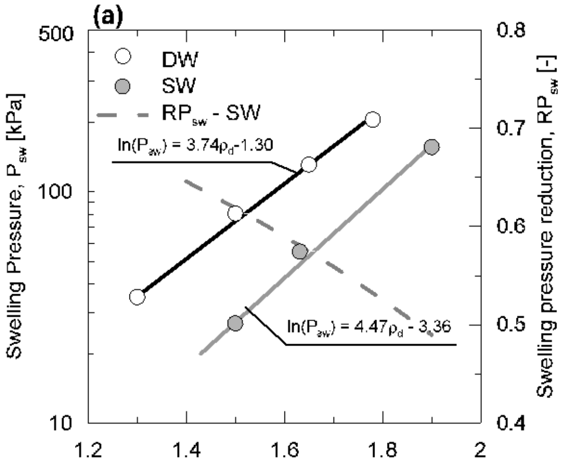
3 EXPERIMENTAL RESULTS

3.1 *Macroscopic swelling behaviour*

The results of the swelling pressure tests performed on the S/B mixture using distilled, synthetic water, and 1 M and 4 M sodium chloride solutions are reported in Figure 3 as a function of the compaction dry density. As expected, the increase in pore water salinity leads to a reduction of the swelling pressure (P_{sw}). In the extreme case of a mixture prepared at the density of 1.8 Mg/m^3 and wetted with the 4 M sodium chloride solution, the swelling pressure was only 15 kPa.

The results of the free swelling tests are synthetized in Figure 4. For all the waters used in the experiments, a linear increase of the swelling deformation (ε_{sw}) with the compaction dry density is observed.

A linear trend was used to fit the swelling strain versus dry density, as suggested by Komine and Ogata (1999). The linear trend would suggest that the minimum dry density required to have an observable macroscopic swelling deformation of the mixture is approximately 1.3 Mg/m^3 when distilled water is used and approximately 1.4 Mg/m^3 when synthetic water is used.



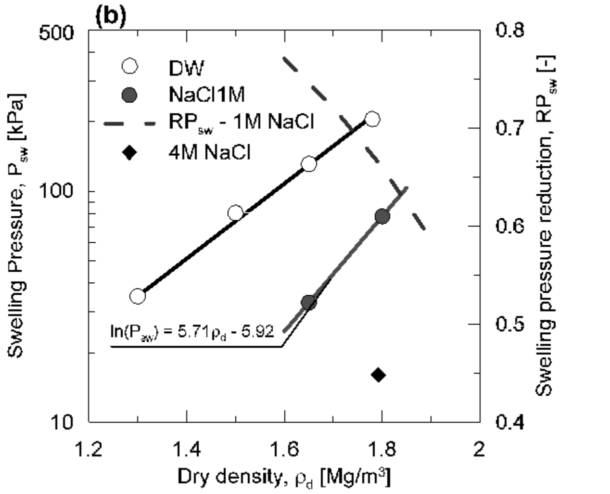


Figure 3. Swelling pressures developed with the different pore waters as a function of the dry density.

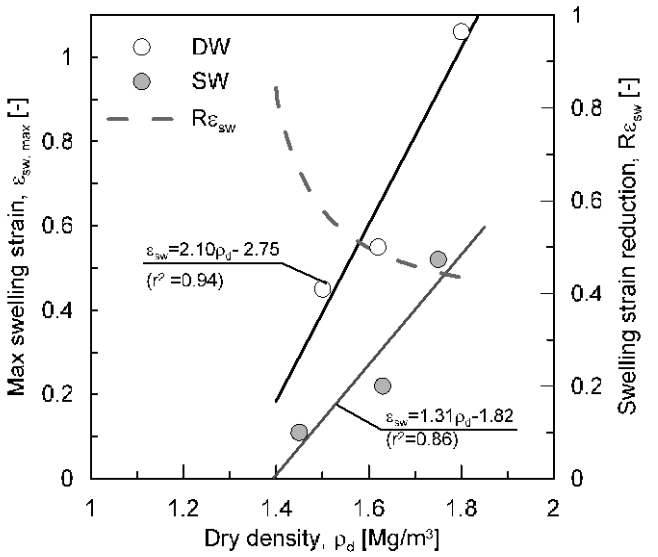


Figure 4. Results of the free swelling tests for distilled and synthetic water.

To quantify the impact of the composition of the pore fluid on the swelling behaviours, the rate of reduction of swelling pressure (RP_{sw}) and the rate of reduction of the free swelling deformation ($R\varepsilon_{sw}$) are depicted in Figure 3 and Figure 4, respectively. The rates of reduction are defined as

$$RP_{sw} = (P_{sw,DW} - P_{sw})/P_{sw,DW} \quad (1)$$

$$R\varepsilon_{sw} = (\varepsilon_{sw,DW} - \varepsilon_{sw})/\varepsilon_{sw,DW} \quad (2)$$

where $P_{sw,DW}$ and $\varepsilon_{sw,DW}$ are the swelling pressure and the maximum swelling strain measured when distilled water was used in the experiments.

The evolution of the swelling pressure with the applied matric suction (s) for two specimens initially compacted to 1.49 Mg/m^3 (e = 0.78) and 1.79 Mg/m^3 (e = 0.49) and wetted with distilled water is presented in Figure 5. The initial matric suction of the mixture at the as-compacted state is approximately 280 kPa, regardless of the compaction density. This indicates that, at the as-compacted state, water is mainly stored in the pores located within the bentonite fraction (Romero et al., 2011). The results show that the swelling pressure is immediately generated with the decrease in matric suction following a non-linear trend in the semi-logarithmic plane. The maximum swelling pressure attained in the test is 59 kPa for the low compacted specimen and 167 kPa for the more compacted specimen. The expected maximum swelling pressures are determined from the results of the conventional swelling test (at 0 kPa matric suction) for the final dry density of the tested specimens. The results were in good agreement with the final swelling pressure generated at a 10 kPa suction, confirming the consistency of the obtained data.

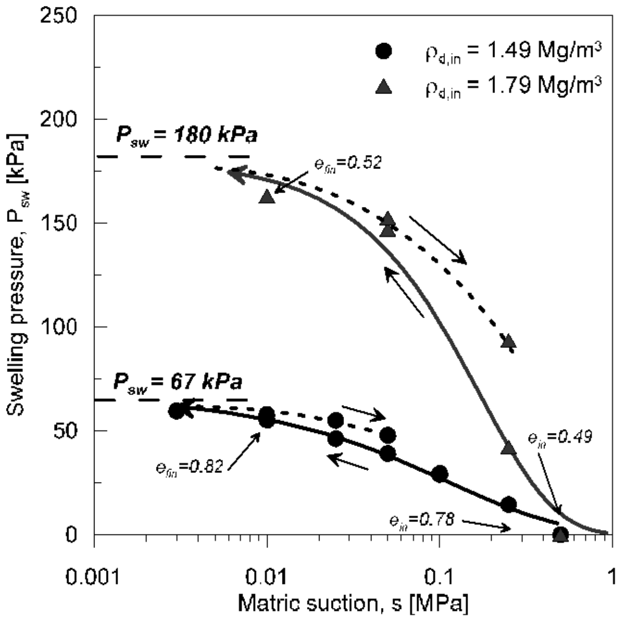


Figure 5. Evolution of the swelling pressure during a wetting process under constant volume conditions for specimen compacted to two different dry densities.

3.2 *Fabric evolution with HCM loadings*

Regardless of the dry density or the salinity of the water used for the preparation of the mixture, the S/B samples in the as-compacted state presented a bimodal pore size distribution (PSD), typical of structured soils compacted to the dry side of the optimum water content (e.g. Delage et al., 1996; Airò Farulla et al., 2010b). Figure 6 depicts the PSD for samples compacted to a dry density of 1.5 Mg/m^3 and wetted with different types of water. In this study, a threshold value of 5 μm is used to delimit the macropore region from the bentonite pore region (intra assemblage pores).

Figure 6 shows that the intra-assemblage pore mode moves toward smaller apparent diameters when the pore water salinity increases, while the macropore mode is only slightly changed.

MIP tests were carried out on S/B samples subjected to monotonic wetting in isochoric conditions. Corresponding matric suction values are derived from the measurements of the water retention behaviour of the mixture (Manca, 2015), accounting for the dependency of the water retention behaviour on the void ratio (Airò Farulla et al., 2010a; Salager et al., 2013). The results are depicted in Figure 7 for a sample compacted to 1.8 Mg/m^3 of dry density. The hydration leads to a movement of the intra assemblage mode towards larger apparent diameters because of the swelling of the assemblages.

The macropore characteristic mode moves toward smaller apparent diameters, and the associated volume is considerably reduced. At the end of the saturation process, the material presented a single mode PSD indicating that all the voids within the grains of sand are filled with bentonite.

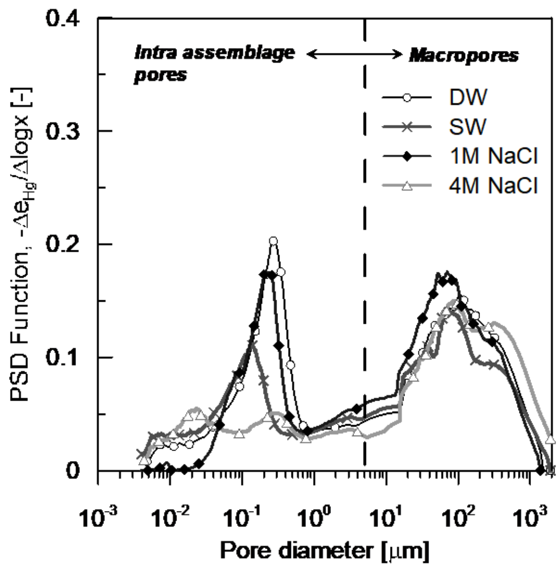


Figure 6. Pore size density functions of samples compacted to 1.5 Mg/m^3 and wetted with different types of water.

Keller et al. (2014) showed that, for the lower dry density, the bentonite wetted with distilled water consists of a gel of very low density arranged in a honeycomb-like structure

The influence of the pore water salinity on the fully saturated mixture is shown in Figure 8, where the PSD are shown for samples compacted to a dry density of 1.5 Mg/m^3 and saturated under constant volume with different types of water. As the pore water salinity is increased, the PSD shows a bi-modal distribution, due to the reduced bentonite swelling at the particle level; these PSDs may determine the inefficiency of the bentonite to properly seal the macropores. As a consequence, the volume and diameters of the macropores are larger when salty water is used.

Further microstructural investigations on the considered S/B mixture are reported in Manca et al. (2016).

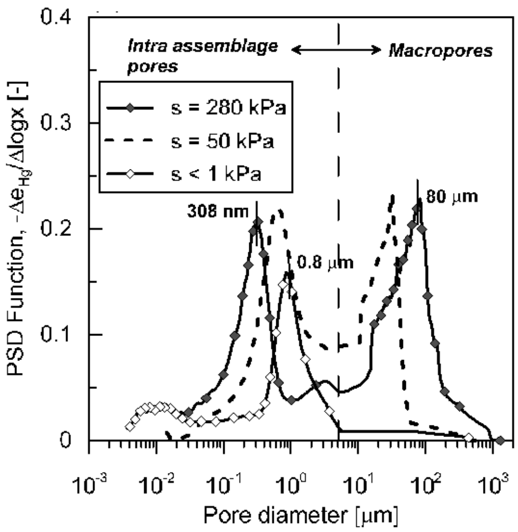


Figure 7. PSD evolution of a S/B mixture subjected to wetting process under constant volume conditions initially compacted to a dry density of 1.8 Mg/m^3.

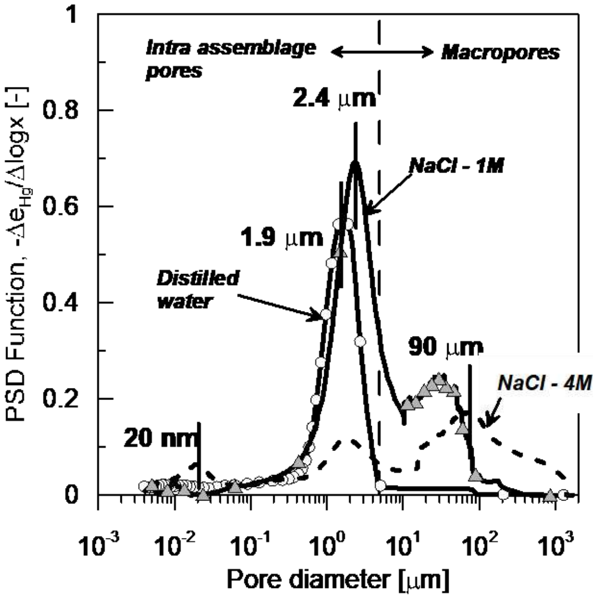


Figure 8. PSD for samples compacted to a dry density of 1.5 Mg/m^3 and saturated under constant volume with different types of water.

4 DISCUSSION

MIP results can be used to analyse the different pore families and to quantify their evolutions with the HCM loadings. Here the void ratio of the S/B mixture is expressed as

$$e = e_M + e_b \tag{3}$$

where e_M is the macrostructural void ratio and e_b is the void ratio of the bentonite aggregates (see Manca et al. (2016) for further details on the computation of the two terms). The bentonite void ratio (e_b) in equation (3) refers to the total volume of solids (bentonite and sand). The relative bentonite void ratio (the ratio of the volume of voids within the bentonite to the bentonite solid volume) and the relative bentonite dry density (the ratio of the dry mass of the bentonite to its total volume) can be computed as follows

$$e_b^{rel} = (e_b \cdot \rho_{s,b}) \,/\, (R_b \cdot \rho_{s,b}) \tag{4}$$

$$\rho_{d,b}^{rel} = \rho_{sb} \,/\, (1 + e_b^{rel}) \tag{5}$$

where $\rho_{s,b}$ is bentonite particle density and R_b is the ratio of bentonite in the mixture. The use of the relative dry density of the bentonite is a useful tool to compare data on the swelling of different bentonite-based materials prepared at different densities (e.g. Agus & Schanz, 2008). Figure 9 reports the swelling pressure of the samples saturated with distilled water as a function of the relative dry density of the bentonite attained at the end of the swelling process. Data from other studies on pure MX-80 bentonite and mixtures containing MX-80 are also reported. A unique trend is observed for the final swelling pressure

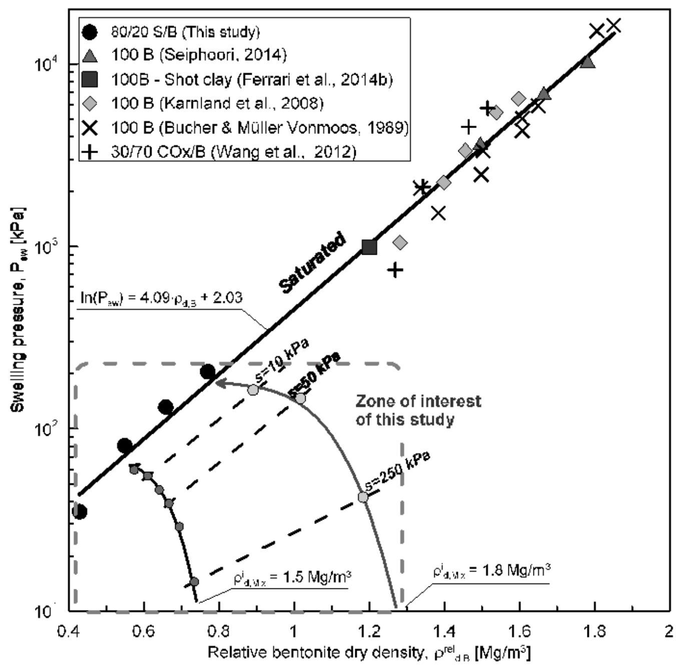


Figure 9. Swelling pressure versus relative bentonite dry density for different soil/bentonite mixtures.

Figure 9 also depicts the results of the suction-controlled wetting tests reinterpreted in terms of relative bentonite dry density (see Manca et al. 2016 for the details). The plot allows to evaluate the generation of the swelling pressure with the decrease in suction associated with the reduction of the relative bentonite dry density. The final swelling pressures of the controlled-suction tests are in very good agreement with the final unique trend.

5 CONCLUSIONS

The paper presented the results of an investigation on the effect of different hydro-chemo-mechanical loadings on the swelling behaviour of a S/B mixture.

The microstructural analyses aided the interpretation of the macroscopic swelling of the S/B mixture. The results revealed that the S/B mixture exhibits a relatively low swelling capacity with respect to other bentonite-based materials prepared with higher bentonite contents. Swelling pressure was found to be suction dependent and very sensitive to chemical loadings.

6 ACKNOWLEDGEMENTS

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7 REFERENCES

Agus, S.S. & Schanz, T. 2008. A method for predicting swelling pressure of compacted bentonites. Acta Geotechnica 3, No. 2, 125–137.

Airò Farulla, C. & Ferrari, A. 2005. Controlled suction oedometric tests: analysis of some experimental aspects. In Proceedings of international symposium on advanced experimental un- saturated soil mechanics, Trento, Italy (eds A. Tarantino, E. Romero and Y. J. Cui), pp. 43–48. London, UK: Taylor & Francis Group.

Airò Farulla, C., Battiato, A. & Ferrari, A. 2010a. The void ratio dependency of the retention behaviour for a compacted clay In Proceedings of 5th International conference on unsaturated soils (Alonso, E., and Gens, A. (eds)). CRC press, Taylor and Francis Group., pp. 417-422.

Airò Farulla, C., Ferrari, A. & Romero, E. 2010b. Volume change behaviour of a compacted scaly clay during cyclic suction changes. Canadian Geotechnical Journal 47(6):688-703.

Bucher, F. & Müller-Vonmoos, M. 1989. Bentonite as a containment barrier for the disposal of highly radioactive wastes. Appl. Clay Sci. 4, No. 2, 157–177.

Castellanos, E., Villar, M., Romero, E., Lloret, A. & Gens, A. (2008). Chemical impact on the hydro-mechanical behaviour of high-density FEBEX bentonite. Phys. Chem. Earth, Parts A/B/C 33, Supplement 1, S516–S526.

Delage, P., Audiguier, M., Cui, Y. & Howat, M. 1996. Microstructure of a compacted silt. Canadian Geotechnical Journal 33(1):150-158.

Ferrari, A., Favero, V., Marschall, P. & Laloui, L. 2014a. Experimental analysis of the water retention behaviour of shales. Int. J. Rock Mech. Mining Sci. 72, 61–70.

Ferrari, A., Seiphoori, A., Rüedi, J. & Laloui, L. 2014b. Shotclay MX-80 bentonite: an assessment of the hydromechanical behaviour. Engng Geol. 173, 10–18.

Karnland, O., Nilsson, U., Weber, H. & Wersin, P. 2008. Sealing ability of Wyoming bentonite pellets foreseen as buffer material–laboratory results. Phys. Chem. Earth, Parts A/B/C 33, Supplement 1, S472–S475.

Keller, L. M., Seiphoori, A., Gasser, P., Lucas, F., Holzer, L. & Ferrari, A. 2014. The pore structure of compacted and partly saturated MX-80 bentonite at different dry densities. Clays Clay Miner. 62, No. 3, 174–187.

Komine, H. & Ogata, N. 1999. Experimental study on swelling characteristics of sand-bentonite mixture for nuclear waste disposal. Soils and Foundations 39(2):83-97.

Komine, H., Yasuhara, K. & Murakami, S. 2009. Swelling characteristics of bentonites in artificial seawater. Can. Geotech. J. 46, No. 2, 177–189.

Manca, D. 2015. Hydro-chemo-mechanical characterisation of sand/bentonite mixtures, with a focus on the water and gas transport properties. PhD dissertation, EPFL, Lausanne, Switzerland.

Manca, D. Ferrari, A. & Laloui L. 2016. Fabric evolution and the related swelling behaviour of a sand/bentonite mixture upon hydro-chemo-mechanical loadings, Géotechnique, 66(1), 41-57.

Nagra 2008. Effects of post-disposal gas generation in a repository for low- and intermediate-level waste sited in the Opalinus Clay of Northern Switzerland, technical report 08-07. Wettingen, Switzerland: Nagra.

Pusch, R. 2001. Experimental study of the effect of high porewater salinity on the physical properties of a natural smectitic clay, technical report 01-07. Stockholm, Sweden: SKB Swedish Nuclear Fuel and Waste Management

Studds, P., Stewart, D. & Cousens, T. 1998. The effects of salt solutions on the properties of bentonite–sand mixtures. Clay Miner. 33, No. 4, 651–660.

Romero, E., Della Vecchia, G. & Jommi, C. 2011. An insight into the water retention properties of compacted clayey soils. Géotechnique 61(4):313-328.

Salager, S., Nuth, M., Ferrari, A. & Laloui, L. 2013. Investigation into water retention behaviour of deformable soils. Canadian Geotechnical Journal 50(2):200-208.

Seiphoori, A. 2014. Thermo-hydro-mechanical characterisation and modelling of MX-80 granular bentonite. Thesis, Ecole Polytechnique Federale de Lausanne (EPFL), Lausanne, Switzerland, Thèse no. 6159.

Seiphoori, A., Ferrari, A. & Laloui, L. 2014. Water retention behaviour and microstructural evolution of MX-80 bentonite during wetting and drying cycles. Géotechnique 64, No. 9, 721–734.

Wang, Q., Tang, A. M., Cui, Y. J., Delage, P. & Gatmiri, B. 2012. Experimental study on the swelling behaviour of bentonite/claystone mixture. Engng Geol. 124, 59–66.

Witteveen, P., Ferrari, A. & Laloui, L. 2013. An experimental and constitutive investigation on the chemo-mechanical behaviour of a clay. Géotechnique 63, No. 3, 244–255, http://dx.doi.org/ 10.1680/geot.SIP13.P.027.