Effect of Suction Changes on the Microstructure of Compacted Crushed Argillites under Constant-Volume Conditions

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Abstract. Crushed and compacted argillite extracted from Bure site (France) has been proposed as a possible sealing and backfilling material in the geological high-level radioactive waste disposal. In order to better understand its coupled hydro-mechanical behaviour in the real storage situation, changes in microstructure of two crushed argillites of different grain size distributions with decreasing suction under confined condition were investigated using mercury intrusion porosimetry (MIP). The results show that wetting under constant-volume condition reduces the size of macro-pores, without significantly affecting the micro-pores size. A clear effect of grain size distribution on the microstructure was also observed: two pore families were identified for the coarse soil; however, there was only one maro-pore family for fine soil (inter-aggregates pores).

Keywords: suction, microstructure, constant volume, MIP, pore size distribution.

1 Introduction

In most conceptual designs of the deep geological repository for high-level radioactive wastes, expansive clays are considered as engineering barrier materials,

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Yu-Jun Cui Ecole des Ponts ParisTech, Paris, France e-mail: yujun.cui@enpc.fr thanks to their low permeability, high swelling and high radionuclide retardation capacities (Pusch, 1979; Yong et al., 1986; Villar et al., 2008). In France, the crushed Callovo-Oxfordian (COx) argillite excavated from the Bure site of the ANDRA underground research laboratory - URL (-490 m, North-eastern France) was proposed as a possible sealing and backfill material, because of the following advantages: (i) it is more economical compared to the commercial bentonites to use local excavated argillite; (ii) the negative impacts on the environment is reduced by recycling the excavated material; (iii) there is better compatibility with host rock because of the same mineralogical and chemical compositions (Andra, 2005; Tang et al., 2011a; Tang et al., 2011b).

In the real repository, after the local groundwater redistribution the water in the host rock formation will move into the repository. Due to the restriction of the host formation, the clays cannot swell, resulting in the development of swelling pressure. The generated swelling pressure will then result in the soil microstructure changes. A good understanding of this micro-structure change is essential as it is directly related to the macroscopic engineering behaviour and physical properties of this material (Cui et al., 2002; Delage, 2007; Tang et al., 2011a).

This study focuses on the compacted crushed Callovo-Oxfordian (COx) argillite. Soils with two different grain size distributions were compacted, and then hydrated by different suctions (from 57 to 38 MPa) using the vapour equilibrium technique under constant volume conditions. After equilibrium, the microstructure was observed using mercury intrusion porosimetry (MIP), allowing the analysis of the effect of suction on the changes of micro-structure.

2 Materials and Experimental Methods

Callovo-Oxfordian (COx) argillite taken from the Bure site of the ANDRA URL was studied. It contains 40–45% clay minerals (mainly interstratified minerals illite–smectite), 20–30% carbonates and 20–30% quartz and feldspar (Hoteit et al., 2000; Lebon & 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/m3 and the specific gravity is 2.70 (Fouché et al., 2004; Tang et al., 2011a).

The excavated argillite was air-dried and crushed following two different procedures, leading to two soils: (1) the coarse soil with the maximum grain size of 8 mm; its air-dried water content was 2.8%; (2) the fine soil with the maximum grain size of 0.25 mm; its air-dried water content was 2.4%. The grain size distributions of the two soils determined by dry sieving are presented in Fig.1; the coarse soil contains 18% fine grains (0.08 mm) while the fine soil contains 70% fine grains. The grain size distribution determined by sedimentation is also presented in Fig.1, the two curves are similar, and it confirms that about 40% grains are clay particles (< 2 μ m).



Fig. 1. Grain size distribution of crushed COx argillite.

MIP tests were performed on samples hydrated with different suctions under constant-volume conditions to analyse the soil microstructure changes with suction. The air dried soil was firstly statically compacted to a dry density of 2.0 Mg/m3 (e = 0.35), with an initial suction of about 150 MPa (measured by a relative humidity sensor). Then, the sample was hydrated under constant volume conditions at different suctions using the vapour equilibrium technique. After equilibrium, the sample was cut to small sticks and freeze-dried. Mercury intrusion was then performed by progressively increasing mercury pressure, and the volume of the pores penetrated during the increase in pressure is continuously monitored (see Delege & Lefevbre, 1984; Cui et al., 2002., Tang et al., 2011a for more details).

3 Results and Discussion

The results of the coarse soil at different suctions are presented in Fig. 2. It is observed that the final values of intruded mercury void ratio (e_m) are lower than the soil void ratio (e = 0.35). This is mainly attributed to the limited range that the MIP technique can cover. For the high-plasticity soils, there is a significant pore volume (entrance diameter smaller than 6 nm) that the mercury could not penetrate to (Delage et al., 2006). The same phenomenon was noted by Lloret et al. (2003) in the pore size distribution curve of compacted Almeria clay (Spain), due to an important amount of pores of very small entrance diameter; the total intraaggregated void ratio appears to be much larger than the intruded one. Comparison of the curves at three suctions shows that with decreasing suction, (150 MPa, 57 MPa, and 38 MPa), the amount of accessible porosity decreases, indicting that during wetting (suction decrease) under constant-volume conditions, the macropore is filled by small clay particles (Cui et al., 2002).

Fig. 3 presents the pore size distribution curves, $de_m/dlog(d)$ versus log(d), of the coarse soil for different suctions. At the initial state (s = 150 MPa, $\rho_d = 2.0 \text{ Mg/m}^3$, e = 0.35), two pore populations are observed, that are micro-pores (having a mean size of 0.03 µm) and macro-pores (having a mean size of 1.5 µm)

(Fig.3). The decrease of suction leads to a slight change in micro-pores size, but a significant change of the macro-pores family (Fig.3): the application of a suction of 57 MPa decreases the modal size of macro-pores to $1.0 \,\mu\text{m}$; a subsequently decrease of suction to 38 MPa reduces the macro-pores size to $0.8 \,\mu\text{m}$. Similar phenomenon was also observed by Cui et al. (2002) who investigated the micro-structure changes of Kunigel/sand mixture with suction under constant-volume conditions. The results show the decrease in inter-aggregate pore size with decreasing suction. It confirms the conclusion that wetting under constant volume conditions leads to clay aggregate hydration, and exfoliation phenomenon occurs around the macro-pores leading to the clogging of macro-pores.



Fig. 2. Intruded mercury void ratio versus pore size for coarse soil.



Fig. 3. Pore size distribution curves of coarse soil at different suction.

The results obtained on the fine soil are presented in Fig. 4 and Fig. 5. In the e_m - log (d) plot (Fig. 4), a lower value of e_m compared to the global void ratio of the soil (e = 0.35) was obtained due to the limited range of the MIP technique as for the coarse soil; with decreasing suction, the amount of accessible porosity decreases as the macro-pore is filled by small clay particles. However, in the de_m/dlog(d) versus log(d) plot (Fig. 5), different phenomenon was observed for the fine soil: there is only one pore family with a mean size of 0.4 µm at the initial state (s = 150 MPa), that also corresponds to the macro-pores family (interaggregates pores), the micro-pores (intra-aggregates pores) are not clearly observed. It indicts the obvious effect of grain size distribution on the microstructure: for the finer soil, the intra-aggregate pores would be hidden by the inter-aggregate

pores having similar size because of the large amount of fine grains created during crushing (see Fig.1). With the subsequent decrease of suction, the macro-pores size reduces to 0.2 μ m and 0.15 μ m at suction of 57 MPa and 38 MPa (Fig.5) respectively, showing the clogging of macro-pores upon wetting.



Fig. 4. Intruded mercury void ratio versus pore size for fine soil.



Fig. 5. Pore size distribution curves of fine soil at different suction.

4 Conclusion

In order to better understand the coupled hydro-mechanical behaviour of the compacted expansive soil used as sealing and backfill material in the deep geological repository for high level radio-active wastes, the microstructure changes of crushed COx argillites (having two different grain size distributions) with decreasing suction under confined conditions are investigated using mercury intrusion porosimetry (MIP). The results show that under constant-volume conditions, decreasing suction reduces the size of macro-pores, but the changes in micro-pores size being not significant. A clear effect of grain size distribution on the microstructure was also observed, two pore families (macro-pores and micro-pores) were identified for coarse soil. By contrast, there was only one pore family for the fine soil (macro-pores) because of the large amount of fine grains produced during crushing.

References

- Andra: Référentiel des matériaux d'un stockage de déchets à haute activité et à vie longue Tome 4: Les matériaux à base d'argilites excavées et remaniées. Rapport Andra No. CRPASCM040015B (2005)
- Cui, Y.J., Loiseau, C., Delage, P.: Microstructure changes of a confined swelling soil due to suction controlled hydration. In: Unsaturated Soils: Proceedings of the Third International Conference on Unsaturated Soils, UNSAT 2002, Recife, Brazil, March 10-13, p. 593. Taylor & Francis (2002)
- Delage, P., Marcial, D., Cui, Y.J., Ruiz, X.: Ageing effects in a compacted bentonite: a microstructure approach. Geotechnique 56(5), 291–304 (2006)
- Delage, P.: Microstructure features in the behaviour of engineered barriers for nuclear waste disposal. In: Springer Proceedings in Physics, vol. 112, p. 11 (2007)
- Fouché, O., Wright, H., Le Cléac'h, J.M., Pellenard, P.: Fabric control on strain and rupture of heterogeneous shale samples by using a non-conventional mechanical test. Applied Clay Science 26(1-4), 367–387 (2004)
- Hoteit, N., Ozanam, O., Su, K.: Geological Radioactive Waste Disposal Project in France: Conceptual Model of a Deep Geological Formation and Underground Research Laboratory in Meuse/Haute-Marne Site. In: The 4th North American Rock Mechanics Symposium, Seattle, July 31-August 3 (2000)
- Lebon, P., Ghoreychi, M.: French Underground Research Laboratory of Meuse/Haute-Marne: THM Aspects of Argillite Formation. In: EUROCK 2000, Aachen, pp. 27–31 (2000)
- Lloret, A., Villar, M.V., Sanchez, M., Gens, A., Pintado, X., Alonso, E.E.: Mechanical behaviour of heavily compacted bentonite under high suction changes. Géotechnique 53(1), 27–40 (2003)
- Pusch, R.: Highly compacted sodium bentonite for isolating rock-deposited radioactive waste products. Nucl. Technol, United States 45(2), 153–157 (1979)
- Tang, C.S., Tang, A.M., Cui, Y.J., Delage, P., Barnichon, J.D., Shi, B.: A study of the hydro-mechanical behaviour of compacted crushed argillite. Engineering Geology 118(3-4), 93–103 (2011a)
- Tang, C.S., Tang, A.M., Cui, Y.J., Delage, P., Schroeder, C., De Laure, E.: Investigating the Swelling Pressure of Compacted Crushed-Callovo-Oxfordian Argillite. Physics and Chemistry of the Earth (special issue) (2011b) (in press)
- Villar, M.V., Lloret, A.: Influence of dry density and water content on the swelling of a compacted bentonite. Applied Clay Science 39(1-2), 38–49 (2008)
- Yong, R.N., Boonsinsuk, P., Wong, G.: Formulation of backfill material for a nuclear fuel waste disposal vault. Canadian Geotechnical Journal 23(2), 216–228 (1986)
- Zhang, C.L., Rothfuchs, T.: Experimental study of the hydro-mechanical behaviour of the Callovo-Oxfordian argillite. Applied Clay Science 26, 325–336 (2004)