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Agustín Molinero Guerra[1,3], Yu-Jun Cui[1], Yong He[2], Pierre Delage[1], Nadia Mokni[3], Anh Minh Tang[1], Patrick Aimedieu[4], Michel Bornert[1], Frédéric Bernier[5]

[1]Ecole des Ponts ParisTech, Laboratoire Navier, France
[2]Central South University, School of Geosciences and Info-Physics, China
[3]Institut de Radioprotection et de Sûreté Nucléaire (IRSN), France
[4]Laboratoire Navier, UMR 8205, ENPC, IFSTTAR, CNRS, UPE, France
[5]Agence Fédérale de Contrôle Nucléaire (AFCN), Belgium

Corresponding author:

Prof. Yu-Jun CUI
Ecole des Ponts ParisTech
6-8 av. Blaise Pascal, Cité Descartes, Champs-sur-Marne
F-77455 Marne – la – Vallée cedex - France
Telephone: +33 1 64 15 35 50
Fax: +33 1 64 15 35 62
E-mail: yu-jun.cui@enpc.fr

Abstract:

The water retention, compressibility and swelling properties of a pellet/powder bentonite mixture were investigated in the laboratory. The water retention capacity was determined under constant-volume condition using a specially designed cell allowing vapor exchange in all directions; suction controlled oedometer tests were carried out for the compressibility investigation; both constant-volume and swell-consolidation methods were applied for the swelling pressure determination. The vapor equilibrium technique was used for suction control. Comparable results were found for the water retention capacity of the pellet/powder mixture determined under constant-volume condition and one single pellet under free swelling condition for suctions higher than 4 MPa, suggesting that the suction was mainly controlled by the micro-pores inside the pellets/powder grains and the swelling of the clay particles was accommodated by the existing macro-pores. On the contrary, at lower suctions, the constant-volume conditions gave rise to a lower water retention capacity for the mixture, indicating the restricted clay particle swelling after the filling-up of the existing macro-pores. Oedometer compression curves revealed a different behavior for low and high suctions: at high suctions, the volume change behavior was governed by the pellets rearrangement combined with possible pellets crushing upon loading. By contrast, at low suctions, the mixture lost its initial granular structure during suction decrease; thus, the volume change behavior became similar to that of compacted bentonite. Compared to compacted bentonite, the pellet/powder mixture showed a lower yield stress. A lower value of swelling pressure was found with the constant-volume method, confirming the limitation of the swell-consolidation method in determining soil swelling pressure.

Keywords: pellet/powder bentonite mixture; laboratory testing; initial heterogeneity; water retention; swelling; compressibility

1. Introduction

The design of most deep geological repositories for high level radioactive wastes (HLW) is based on the multi-barrier concept which involves the natural host rock and engineered barriers (Sellin and Leupin, 2013). In this design concept, bentonite-based materials are used to construct buffers, backfills and seal plugs of disposal galleries and access shafts to ensure isolation of the waste from the environment. In particular, mixtures made up of bentonite powder and pellets are considered as a promising candidate sealing material because of its particular properties. In addition to their low permeability, high swelling capacity and high radionuclide migration retardation properties, the pellet/powder mixture exhibits operational advantages in terms of emplacement and avoids the formation of technological gaps between the host rock and the seal plug. Once installed in the repository, the pellet/powder mixture will undergo both hydraulic and mechanical loadings: infiltration of host rock pore water and mechanical confinement imposed by the surrounding rocks. In this context, to ensure the stability of the storage structure, thus the safety of the disposal system, it is of paramount importance to well understand the hydro-mechanical behavior of unsaturated bentonite pellet/powder mixture. For this purpose, the French Institute for Radiation Protection and Nuclear Safety (IRSN) launched the SEALEX project, which consisted of a series of in situ experiments in the Underground Research Laboratory (URL) in Tournemire, France (Mokni and Barnichon, 2016; Mokni, 2016). This project aimed at investigating the long-term hydraulic performance of sealing systems in normal and critical scenarios, with different sealing plug configurations (pre-compacted or field compacted sand/bentonite mixtures, pellet/powder bentonite mixtures). In parallel to the in situ experiments, mock-up tests were also carried out on compacted sand/bentonite blocks (Wang et al., 2012; Saba et al., 2014; Mokni, 2016). The swelling pressure of compacted sand/bentonite blocks was also determined by Wang et al.

(2013a). The results confirmed the dominating role of the block dry density, identified by many researchers (Börgesson et al., 1996; Dixon et al., 1996; Lloret et al., 2003; Villar, 2005; Gens et al., 2011; Villar et al., 2012; Schanz and Al-Badran, 2014). Further examination showed that sand in the mixture did not contribute to the swelling pressure; it was the dry density of bentonite contained in the mixture that defined the final swelling pressure of the sand/bentonite mixture (Wang et al., 2012; Saba et al., 2014), in agreement with the results obtained by previous researchers (e.g. Villar and Rivas, 1994; Karnland et al., 2008). The well-established relationship between final swelling pressure and bentonite dry density constituted a helpful tool in interpreting the field observation in the SEALEX project (Wang et al., 2013a).

As opposed to the compacted bentonite-based blocks, the pellet/powder bentonite mixture has been scarcely investigated in the laboratory. Hoffmann et al. (2007) performed different tests on mixtures of different-size pellets of Febex bentonite at different dry densities considered within the Engineered Barrier (EB) project. An important finding was the progressive decrease of water permeability induced by the clogging of the large inter-pellet pores due to granule swelling and the development of both swelling strain and pressure upon wetting. Imbert and Villar (2006) investigated the hydro-mechanical behavior of a 50/50 FoCa bentonite pellet/powder mixture by a series of infiltration tests. They concluded that the mixture became homogeneous with a swelling pressure equal to that of a bentonite powder compacted at the same dry density once full saturation was reached.

In this study, the water retention capacity and the swelling and compressibility properties of a 80/20 pellet/powder MX80 bentonite mixture were investigated. Firstly, the water retention curve (WRC) was determined by following a wetting path under constant-volume conditions using a specially designed cell with vapor exchange allowed in all directions. This curve was compared to that of a single pellet of bentonite swelling under free swelling conditions. Secondly, a series of suction controlled oedometer tests were performed to investigate the

compressibility properties of the mixture. The swelling behavior of the mixture after following a wetting-drying path was also studied in oedometer. Finally, the swelling pressure was investigated using both constant-volume and swell-consolidation methods. Emphasis was put on the size of samples due to the strong heterogeneous nature of the material with large pellets. The results obtained allowed the main hydro-mechanical properties of the bentonite pellets/powder mixture used in the SEALEX project to be characterized, providing relevant parameters for the numerical analysis of the field infiltration experiments involving this mixture.

2. Materials and methods

2.1. Materials

The investigated material was a mixture of pellet/powder of MX80 bentonite at a proportion of 80/20 in dry mass. The MX80 bentonite came from Wyoming, USA, and had a smectite content of 80%, other non-clayey inclusions being quartz, calcite and pyrite. The cation exchange capacity (CEC) was 98 meq/100g, with Na^+ as major exchangeable cation, with a value of 52 meq/100g (1.2 meq/100g for K^+, 10 meq/100g for Mg^{2+} and 37 meq/100g for Ca^{2+}). The liquid limit was 560%, the plastic limit was 62% and the unit mass of solid was 2.77 Mg/m^3 (Saba et al., 2014).

Pellets were produced in the Laviosa-MPC company by instantaneously compacting MX80 bentonite in a mould of 7 mm in diameter and 7 mm in height. A zoom on a single pellet is shown in Figure 1. The fabrication water content was $w = 6.0 \pm 1.0\%$ and the dry density was $\rho_d = 2.06 \pm 0.06$ Mg/m^3, corresponding to a void ratio $e = 0.30 \pm 0.07$. The pellets were stored in the laboratory in a hermetic plastic box at 20°C. The initial suction ($s = 135 \pm 3$ MPa) was measured in the laboratory with a chilled mirror dew point hygrometer (Decagon WP4C), at an initial water content $w = 7.2 \pm 1.0\%$ (determined after oven-drying at 105°C), slightly higher

than the fabrication one due to the water adoption from air after fabrication (Molinero Guerra et al., 2017). Note that the WP4C measures corresponded to total suction, including the osmotic suction.

The MX80 bentonite powder was produced by crushing pellets just after their fabrication. The maximum size of grains was 2 mm and the portion of the grains smaller than 80 μm was 5%. An initial water content of 3.2 ± 1.0% was found in the laboratory by oven-drying at 105°C for 24 h, corresponding to an initial suction s = 191 MPa (measured with WP4C). This water content was quite low compared to the fabrication water content (6.0 ± 1.0%), suggesting loss of some water during the pellets crushing operation. More details about the initial state of the material can be found in Molinero Guerra et al. (2017).

2.1. Sample preparation

All the results presented in this study were obtained by performing laboratory tests on the pellet/powder bentonite mixture. The mixture was prepared by following the first protocol presented in Molinero Guerra et al. (2017) to obtain a relatively homogeneous pellet/powder distribution. It consisted in filling the cell by packets corresponding to one layer of pellets spread over the base of the cylinder, with the corresponding amount of powder, respecting the proportion of 80-pellets/20-powder. The global dry initial density of the mixture thus prepared was 1.49 Mg/m^3.

In order to better understand the initial pellet/powder distribution, microfocus X-ray computed tomography observations were made on the mixture at its initial state. A vertical slice is presented in Figure 2. The dimensions of the observed sample were 120 mm in height and 60 mm in diameter. It appears that even though a special protocol was followed, it was difficult to perfectly control the homogeneity of the sample. Indeed, the grains of powder were not uniformly distributed within the sample, and inter-pellet voids could be clearly identified. The

sample, placed between two porous stones, presented larger inter-pellet voids close to the top porous stone and no grains of powder were observed in this zone. Therefore, this material was characterized by a strong heterogeneity at its initial state, which is an important aspect to take into account while further analyzing the global hydro-mechanical behavior of the sample. This initial heterogeneity was investigated in depth by Molinero Guerra et al. (2017).

The samples used for the present study were prepared in the same manner, but with a height of 35 mm. This height corresponded to 5 times the height of a single pellet and it was believed that such dimensions constituted a representative volume of the pellet/powder mixture. This ratio of sample size to maximum grain size was also adopted by Duong et al. (2013) when investigating the mechanical behavior of a mixture of coarse and fine soils.

2.2. Experimental methods

2.2.1. Water retention properties

The water retention curve (WRC) of the pellet/powder bentonite mixture under constant-volume conditions was determined on samples of 50 mm in diameter. A special device presented in Figure 3 was designed for this purpose. The pellet/powder mixture was placed into a rigid stainless steel cell that was designed to allow vapor exchanges not only through the two metallic porous disks on both top and bottom of the sample, but also through a cylindrical porous disk on the lateral surface of the sample. This design was expected to significantly accelerate the hydration of the sample by vapor equilibrium technique, essential for low-permeability materials like the studied swelling MX80 bentonite. The mass of sample was regularly measured. In a standard fashion, equilibrium was considered once the mass changes became insignificant. For each pellet/powder mixture, once equilibrium was reached, three measures were performed for both the water content by oven drying and the suction with WP4C. The mean values were considered in the further analyses.

178 In parallel, the water retention curve under free swelling conditions for a single pellet of
179 bentonite was obtained. To this end, pellets were hydrated under several values of suction from
180 their initial state by vapor equilibrium technique. For the pellet/powder mixture, zero suction
181 was applied by injecting deionized water into the cell under a low pressure corresponding to a
182 water column height of about 200 mm.

183 *2.2.2. Compressibility properties – suction controlled oedometer tests*

184 Controlled suction oedometer compression tests were performed on samples of 35 mm in height
185 and 50 mm in diameter by circulating vapor at controlled relative humidity through the base of
186 the sample as shown in Figure 4 (tests SCO1, SCO2 and SCO3 presented in Table 1). In order
187 to investigate the mechanical behavior of the mixture in a relatively large range of load, a high
188 pressure oedometer was used allowing application of vertical stresses as high as 50 MPa
189 (Marcial et al., 2002). This oedometer was previously calibrated using a force transducer in
190 order to ensure the accuracy of the imposed pressures. As opposed to other suction values, zero
191 suction was imposed by circulating deionized water. Vertical displacement was monitored
192 using a digital micrometer (accuracy ± 0.001 mm). The temperature in the laboratory was
193 controlled at 20 ± 0.5°C during the experiments.

194 Time was allowed until swelling stabilized under the imposed value of suction and a low
195 vertical stress of 0.1 MPa. The stabilization was considered when the vertical displacement was
196 lower than 0.01 mm for, at least, 8 h. It is worth noting this criterion was defined based on the
197 French Standard AFNOR (1995) for oedometer compression test. The real swelling
198 stabilization might take much longer time. Three tests were carried out under three different
199 suctions (0 MPa, 9 MPa and 138 MPa). The test at 138 MPa suction (SCO1) was performed
200 without imposing the suction, assuming that the suction changes during loading-unloading were
201 negligible. In this case, the test was performed at constant water content. This assumption was
202 supported by the results of Gens et al. (1995) who measured the suctions of a soil compacted

under three different stresses and observed that on the dry side of optimum the soil suction was almost the same, suggesting that inter-aggregate macro-pores were dry. Similarly, in this study, the macro-pores of the pellets/powder mixture could be considered as dry at a suction as high as 138 MPa. Thus, it is assumed that compression decreased the macro-pores but not the suction of the mixture.

Loadings were performed in steps from 0.1 MPa. A new load equal to the double of the previous one was applied (except the last loading step for sample SCO1) when the change of the vertical displacement became insignificant (< 0.01mm for 8 h according to AFNOR, 1995). The maximums loads were 12.8 MPa for SCO2, SCO3 and SCO4 and 18 MPa for SCO1 because of the higher stiffness of this latter due to its higher suction. After the loading process, unloadings were performed in steps down to different load values.

2.2.3. Swelling properties

Three tests, SCO3, SCO4 and CV1 presented in Table 1, were performed in order to study the swelling properties of the pellet/powder bentonite mixture. Both the constant-volume and swell-consolidation methods were applied to determine the swelling pressure. To apply the constant-volume method, a constant volume cell was used, which included three parts: (i) the bottom part which contained a porous stone and a drainage system; (ii) the middle cell (70 mm in inner diameter) which prevented any radial swelling, with two air outlets; (iii) the top part including a total pressure sensor for monitoring the axial swelling pressure. The water inlet was connected to a water reservoir placed at the same height of the cell, flooding the pellet/powder mixture from the bottom. The saturation water used was a synthetic water (see Table 2 for the receipt of preparation), which has the same chemical composition as the pore water of the Callovo-Oxfordian claystone in the underground research laboratory of the French agency for radioactive waste management (ANDRA) in Bure, France. In the swell-consolidation method, the sample was hydrated under a low vertical load of 0.1 MPa until full swelling was achieved.

Then, it was compressed following a standard consolidation test procedure. The pressure needed to compress the specimen back to its initial volume was defined as the swelling pressure of the material (Basma et al., 1995; Abdullah and Mhaidib, 1998, 1999; Agus, 2005, Wang et al. 2012).

3. Experimental results

3.1. Water retention curve

Figure 5 displays the evolution of water content with time for the pellet/powder mixture equilibrated at different suctions. The special cell designed allowed the samples to reach water content stabilisation quite quickly. This was because with the specially designed cell (Figure 3), the sample was completely surrounded by porous stones, which accelerated the vapor exchange process. For all imposed suctions, the water content was stabilized after 30 days of hydration, except for 4 MPa and 82 MPa. For s = 4 MPa, some technical problems occurred during the test, and a constant water content value was found after 80 days. It is suspected that, at this low suction, the hydration system failed after a certain time due to water condensation in the pipes – cleaning intervention was necessary to solve the problem. The slight temperature fluctuations of ± 0.5°C in the laboratory during the experiments could cause this condensation. For suction s = 82 MPa, it seems that water content was constant after 30 days; nevertheless, it continued increasing slightly up to 90 days. For this value of suction, several problems were found during wetting in the hydration system (blockage of the vapor circulation system) and regular interventions were needed. It could be considered that this technique ran well for suctions higher than or equal to 9 MPa. For lower suctions, problems related to condensation in the pipes could be significant in the relatively long tests (more than 80 days). These problems are to be solved in further studies, by placing the main system in water with temperature control for example.

252 The water retention curves under constant (pellet/powder mixture) and free swelling conditions
253 (single pellet) for a wetting path are presented in Figure 6 in terms of water content versus
254 suction. It appeared that pellet and mixture had comparable water retention capacities when
255 suction was higher than 4 MPa, suggesting that the suction was governed by the micro-pores
256 within the pellets or powder grains. In addition, in the case of the mixture, the existing macro-
257 pores can accommodate the swelling of the clay particles. This is in agreement with the results
258 obtained by Yahia-Aissa et al. (2001) on compacted Foca 7 clay. It seems that the mixture had
259 a lower water retention capacity under constant-volume conditions for suctions lower than 4
260 MPa, since its water content at zero suction was 34.3%. This indicated the restriction effect on
261 the clay particles swelling, thus the water retention capacity. More data are needed in this range
262 of suction to confirm this conclusion. Figure 7 presents the suctions measured by WP4C versus
263 the imposed values by either vapor equilibrium technique (for non-zero suctions) or injecting
264 deionized water (for zero suction) for the pellet/powder mixture.

265 Some differences were found between the measured suction after equilibrium and the imposed
266 value by vapor equilibrium technique, in particular for zero suction. This could be due to the
267 stress release while dismantling the specimen, which led the suction to increase within the
268 mixture. This effect appeared more pronounced in the case of zero suction due to the highest
269 swelling pressure developed, which enhanced the stress release effect.

270 Pintado et al. (2009) concluded that, with the vapor equilibrium technique, it was difficult to
271 reach the same suction distribution inside the sample. From Figure 7, it appears that the suction
272 imposed and the suction measured were almost the same, indicating that in our case the uniform
273 suction distribution was reached thanks to the additional air flow through the lateral surface.
274 This shows the performance of the designed device.

275 *3.2. Controlled suction compression tests (SCO1, SCO2 and SCO3)*

Figure 8 presents the evolution of volumetric strain with time for tests SCO1, SCO2 and SCO3. The test at 138 MPa of suction was performed without imposing suction. It was carried out at constant water content conditions. As mentioned before, the suction can be reasonably considered as constant in this test.

In test SCO3, the pellet/powder mixture was hydrated under 0.1 MPa by injecting deionized water to impose zero suction. Equilibrium was reached after 1272 h (53 days), with a value of 21% of volumetric strain, corresponding to a void ratio of e = 1.25. The volumetric strain at equilibrium was higher than the value obtained by Wang et al. (2012) for compacted bentonite/sand mixture: 18% (e = 0.97). This could be explained by the higher swelling capacity of the pellet/powder mixture without sand. In test SCO2 for 9 MPa suction applied by vapor equilibrium technique, equilibrium was achieved after 1032 h (43 days), with a volumetric strain of 9% (e = 1.02). A constant-volumetric strain was reached between 320 h and 430 h. This is due to a problem in the hydration system that needed a cleaning intervention. After 430 h, the specimen continued swelling until reaching equilibrium. In test SCO1, no suction was imposed as 138 MPa corresponded to the initial state of the pellet/powder mixture; a period of 5 days was considered prior to loading in this case.

Once equilibrium was reached, the samples were submitted to loading/unloading at constant suctions. For SCO3 (zero suction), on the whole, the evolution of volumetric strain in each loading step showed typical consolidation features of fine-grained soils, in particular under high loads. On the contrary, for SCO1 (138 suction) and SCO2 (9 MPa suction), the evolution curves were rather characterized by the presence of almost instantaneous volumetric strains after each loading. It was thus suspected that when suction was higher than 9 MPa, it was rather the pellets/grains arrangement or crushing that governed the volume change behavior of the mixture.

Test SCO4 was designed to study the volume change behavior upon wetting/drying in oedometer. In this case, the pellet/power mixture was submitted to a wetting-drying cycle at a constant vertical stress of 0.1 MPa. In the first phase, the specimen was hydrated by injecting deionized water during 1248 h (52 days) to reach equilibrium at zero suction (comparable to the 1272 h for test SCO3). A volumetric strain of 23% (e = 1.28) was found at equilibrium. Then, a suction of 262 MPa was imposed to the mixture. The volumetric strain decreased drastically, reaching a value of 5% (e = 0.87) after 1375 h (57.3 days). A final value of 3% (e = 0.84) was observed after 2311 h (96.3 days).

Once equilibrated at the imposed suctions, the pellet/powder mixtures were submitted to compression at constant suctions. The results are displayed in Figure 9 in terms of void ratio with respect to vertical stress. The initial void ratio values were different due to the different swellings under different suctions. In a standard fashion, the compression curves were characterized by an initial linear part with low compressibility (that could be considered as the pseudo-elastic domain) followed by a second part with higher compressibility (that could be considered as the plastic domain). Finally, the third part, corresponding to the unloading, exhibited the same trend as the elastic domain, as only elastic deformations were expected. For test SCO3 where zero suction was applied, the unloading path could also be considered as bi-linear.

The swelling pressure of the pellet/powder mixture could be determined by the swell-consolidation method using the results from test SCO3. As it was explained previously, the swelling pressure of the material corresponded to the vertical stress needed to compress the sample to its initial volume. The obtained value was $\sigma_s = 7.1$ MPa (e = 0.859).

3.3. Swelling under constant-volume conditions (CV1)

323 The swelling behavior of the pellet/powder bentonite mixture under constant-volume
324 conditions was investigated by following the evolution of the axial swelling pressure during
325 hydration (CV1 in Table 1). The results are displayed in Figure 10. Two rates of increase of
326 axial swelling pressure could be distinguished from the beginning of the hydration process: the
327 first one corresponded to a period from the beginning of the hydration up to 20 days, when
328 swelling pressure reached 0.43 MPa. Then, the pressure continued increasing at a different rate,
329 until 45 days of hydration. At this moment, the swelling pressure reached 0.8 MPa. Afterwards,
330 a slight increase was observed, finally reaching 0.87 MPa after 440 days. Nevertheless, the
331 swelling pressure continued increasing, which means that the sample was not completely
332 stabilized, even though the rate of increase was quite low compared to those observed before.
333 The value of pressure observed after 440 days (0.87 MPa) was significantly lower than the
334 value obtained by applying the swell-consolidation method on the consolidation curve (7.1
335 MPa).

336 # Discussion

337 The swell-consolidation and constant-volume method revealed different values of swelling
338 pressure: the swell-consolidation method provided 7.1 MPa, whereas a value of 0.87 MPa was
339 found with the constant-volume method. This was common results and Wang et al. (2012)
340 explained that by the coupling between the microstructure deformation and the macrostructure
341 deformation, combined to the effect of friction.

342 Cui et al. (2013) carried out oedometer tests with loading/unloading/reloading on natural stiff
343 Ypresian clays (YPClay). They concluded that each unloading/reloading curve could be
344 satisfactorily considered as bi-linear with a small and a larger slope separated by a threshold
345 vertical stress. This stress was found, through hydrations tests under constant volume
346 conditions, to be the swelling pressure corresponding to the void ratio just before the unloading

or reloading. By applying this approach to the pellet/powder mixture at zero suction (SCO3), with the consideration of the unloading path as bilinear, a swelling pressure of 4.2 MPa (threshold vertical stress) would be obtained, corresponding to a void ratio $e = 0.72$ (dry density $\rho_d = 1.61$ Mg/m^3). This value was comparable with that estimated by the method of Wang et al. (2012): a value of 4.15 MPa of swelling pressure was found at a dry density of 1.49 Mg/m^3. This showed that the true swelling pressure of the mixture should be lower than that determined by the swell-consolidation method, because the value would correspond to the point separating the two parts of unloading curve, thus below the consolidation curve.

Further analysis of the swelling pressure evolution with time displayed in Figure 10 revealed the processes governing the structural changes of the pellet/powder mixture while wetting. At the very beginning of the test with wetting from the bottom, the observed swelling pressure and the corresponding rate of increase were due to the pellets located at the top of the sample, where the suction was still high. After a period of 20 days, a second rate of increase higher than the first one was observed. The reason was that water has arrived to pellets or powder grains located at the top of the sample at this state; thus, the involved pellets and powder grains started swelling and the pressure increased accordingly. It was suspected that all inter-pellet/grain voids were filled with swollen bentonite after 40 days of hydration, as the rate of increase of swelling pressure decreased. However, the rate was not equal to zero, meaning that the specimen was not homogeneous. Taking into account the relationship proposed by Wang et al. (2013a) between the swelling pressure and the dry density of bentonite, a final value of 4.15 MPa of swelling pressure was found. Obviously, the swelling pressure of 0.87 MPa obtained by the constant method was quite far from this value. Molinero Guerra et al. (2018) performed an infiltration test on the same mixture with the same global dry density (1.49 Mg/m^3) using a metallic column (60 mm in diameter and 120 mm in height) equipped with a force transducer for the axial swelling pressure measurement and several total pressure transducers for the radial

swelling pressure measurements at different positions (20 mm, 40 mm, 60 mm, 80 mm and 100 mm from the bottom of sample). Water infiltration was allowed from both the top and the bottom of sample. They observed an axial swelling pressure of 2.5 MPa after 845-day infiltration, also lower than the expected value (4.15 MPa). The radial pressures at 20 mm, 40 mm, 60 mm, 80 mm were almost the same, around 4 MPa. By contrast, the value at 100 mm was lower, around 3 MPa. In addition, the swelling pressures were developed mainly during the first 100 days, the variations after 100 days appearing negligible. They attributed the difference between the radial swelling pressures by the heterogeneity of the sample: a higher swelling pressure was expected at a position where the local dry density was greater. They explained the lower axial pressure by the effect of friction between the sample and the column wall, as also identified by Saba et al. (2014). Indeed, the expected sample homogenization process implied the local movements of bentonite particles. Thus, the wall friction could be mobilized even in constant-volume condition. The results obtained in this study showed that even for a sample of 35 mm height, the effect of cell wall friction was significant, hampering or slowing down the sample homogenization process.

The results from test SCO4 allowed the reversibility of swelling of the pellet/powder mixture to be investigated. After wetting under zero suction, the sample was dried by imposing s = 262 MPa. The results showed a drastic decrease of volumetric strain, reaching a final value of 3%. This suggested that plastic swelling strain occurred upon wetting under zero suction, as shown in Figure 11. This was probably due to pellets fissuring upon wetting. Wang et al. (2013b) also reported the fissuring phenomenon for compacted bentonite/sand mixture upon wetting. Compared to test SCO3, a slightly higher volumetric strain was reached in test SCO4, suggesting a certain effect of the pellet/powder distribution that is specific for each sample. The final points of tests SCO1, SCO2 and SCO3 appeared well aligned, defining a wetting line. This

line was below the drying line estimated from test SCO4. The difference between the two lines was larger at higher suction.

The evolution of the yield stress with suction is presented in Figure 12, together with the values obtained by Marcial et al. (2003) and Wang et al. (2013b) for compacted bentonite with initial void ratio of 0.559 and compacted bentonite/sand mixture with initial void ratio of 0.635, respectively. A good coincidence was observed for compacted bentonite and the pellet/powder mixture at zero suction. For other values of suction, in a standard fashion, the yield stress increased as suction increased. However, this phenomenon was not clear for the pellet/powder mixture: a much lower yield stress value was obtained. A possible explanation is as follows: for non-zero suctions, the pellet/powder mixture did not completely lose its initial granular mixture; thus, the deformations were essentially governed by the rearrangement and possible crushing of pellets while loading. These deformations were of plastic nature. However, in the case of zero suction, the mixture entirely lost its granular structure; thus, the volume change behavior was comparable to that of a common compacted bentonite. This observation was supported by analyzing the evolution of volumetric strain with time: at 138 MPa and 9 MPa of suctions, the volumetric strains decreased almost instantaneously while loading; however, at zero suction, the decrease rate of volumetric strain was clearly lower than those observed at other suctions, because the pellets lost their granular structure in that case.

4. Conclusion

The water retention, compressibility and swelling properties of a 80-pellet/20-powder bentonite mixture were investigated through the water retention tests under free (pellet) and constant-volume (mixture) conditions, the suction controlled oedometer tests and the hydraulic tests on samples at a dry density of 1.49 Mg/m^3.

419 The new cell designed for the WRC determination under constant-volume condition allowed
420 the vapor exchange in all directions, which significantly accelerated the hydration process.

421 Similar water retention properties were observed for pellet/powder mixture under constant-
422 volume condition and single pellet under free swelling condition at suction higher than 4 MPa,
423 indicating that the suction was mainly controlled by the micro-pores inside the pellets or powder
424 grains, and the swell of clay particles was accommodated by existing macro-pores in the
425 mixture. On the contrary, at lower suctions, constant-volume condition defined a lower water
426 retention capacity, showing the restriction effect on clay particles swelling and thus their
427 hydration. Note that more tests are needed to complete the data in the range of suctions lower
428 than 4 MPa.

429 The investigation of the compressibility properties revealed lower values of yield stress than
430 the common bentonite mixtures, except for the specimen hydrated at zero suction. This is due
431 to the granular structure in the case of non-zero suction where the volume change behavior was
432 suspected to be governed by the rearrangement and crushing of pellets, and the loss of the
433 granular structure in the case of zero suction.

434 The irreversibility of the volumetric strain was evidenced by imposing a wetting-drying cycle
435 to the pellet/powder mixture under constant loading. This was probably due to the pellets/grains
436 fissuring upon wetting.

437 Different swelling pressure values were obtained by applying different methods. A higher
438 swelling pressure was obtained with the swell-consolidation method. Further analysis based on
439 the method proposed by Cui et al. (2013) showed that the swell-consolidation systematically
440 overestimates the swelling pressure because for a given density the swelling pressure must be
441 lower than the corresponding pressure on the consolidation curve: the swelling pressure
442 corresponded to the point separating the two parts of unloading curve, thus below the

consolidation curve. Nevertheless, the swelling pressure determined by the constant-volume method seemed quite low (0.87 MPa) with respect to the expected one (4.15 MPa). This could be attributed to the effect of cell wall friction because the expected sample homogenization process implied the local movements of bentonite particles, thereby mobilizing the cell wall friction even in constant-volume condition and with a sample of limited height (35 mm). The cell wall friction would hamper or slow down the sample homogenization process.

References

Abdullah, I., Mhaidib, A., (1998). Prediction of swelling potential of an expansive shale. Proceedings Of The Second International Conference On unsaturated soils. 27–30 August. Beijing, China, vol. 1.

Abdullah, I., Mhaidib, A., (1999). Swelling behaviour of expansive shales from the middle region of Saudi Arabia. Geotechnical and Geological Engineering 16 (4), 291–307.

AFNOR, (1995). Sols : reconnaissance et essais: essai de gonflement à l'oedomètre, détermination des déformations par chargement de plusieurs éprouvettes. XP P 94-091

Agus, S., (2005). An Experimental study on hydro-mechanical characteristics of com- pacted bentonite-sand mixtures. PhD thesis. Weimar.

Basma, A.A., Al-Homoud, A.S., Husein, A., (1995). Laboratory assessment of swelling pres- sure of expansive soils. Applied Clay Science 9 (5), 355–368.

Börgesson, L., Karnland, O. and Johannesson, L. E., (1996). Modelling of the physical behaviour of clay barriers close to water saturation. Engineering Geology 41, No. 1–4, 127–144.

Cui, Y. J., Nguyen, X. P., Tang, A. M., and Li., X. L., (2013). An Insight into the Unloading/reloading Loops on the Compression Curve of Natural Stiff Clays. Applied Clay Science 83-84: 343–48.

Dixon, D. A., Gray, M. N. and Graham, J., (1996). Swelling and hydraulic properties of bentonites from Japan, Canada and the USA. Environmental Geotechnics 1, 43–48.

Duong, T. V., Tang, A. M., Cui, Y. J., Trinh, V. N., Dupla, J. C., Calon, N., Canou, J., and Robinet, A. (2013). Effects of fines and water contents on the mechanical behavior of interlayer soil in ancient railway sub-structure, *Soils Found.*, vol. 53, no. 6, pp. 868–878.

Gens, A., Alonso, E.E., Suriol, J. & Lloret, A. (1995). Effect of structure on the volumetric behaviour of a compacted soil. Proc. of 1st conference on unsaturated soils. vol. 2, pp. 83-88.

Gens, A., Vallejan, B., Sánchez, M., Imbert, C., Villar, M.V. and Van Geet, M., (2011). Hydro mechanical Behavior of a Heterogenous Compacted Soil: Experimental Observations and Modelling.

Hoffmann, C., Alonso, E.E. & Romero, E., (2007). Hydro-mechanical behaviour of bentonite pellet mixtures. Physics and Chemistry of the Earth, 32(8-14), pp.832–849.

Imbert, C. & Villar, M.V., (2006). Hydro-mechanical response of a bentonite pellets/powder mixture upon infiltration. *Applied Clay Science*, 32(3-4), pp.197–209.

Karnland, O., Nilsson, U., Weber, H., Wersin, P., (2008). Sealing ability of Wyoming bentonite pellets foreseen as buffer material – laboratory results. Physics and Chemistry of the Earth, Parts A/B/C 33, S472–S475.

Lloret, A., Villar, M. V., Sánchez, M., Gens, A., Pintado, X. and Alonso, E. E., (2003). Mechanical behaviour of heavily compacted bentonite under high suction changes. Géotechnique 53, No. 1, 27–40.

Marcial, D., Delage, P., Cui, Y.J., (2002). On the high stress compression of bentonites. Canadian Geotechnical Journal 39, 812–820.

Marcial, D., (2003). Comportement hydromécanique et microstructural des matériaux de barrière ouvragée, thèse. ENPC Paris, France.

Mokni, N. & Barnichon, J.D., (2016). Hydro-mechanical analysis of SEALEX in situ tests-Impact of technological gaps on long term performance of repository seals. Engineering Geology, 205, pp. 81-92.

Mokni, N., (2016). Analysis of hydro-mechanical behaviourbehavior of compacted bentonite/sand mixture using a double structure formulation. Environ Earth Sci (2016) 75: 1087.

Molinero-Guerra, A., Mokni, N., Delage, P., Cui, Y. J., Tang, A. M., Aimedieu, P., Bernier, F. and Bornert, M., (2017). In-depth characterisation of a mixture composed of powder/pellets MX80 bentonite. Applied Clay Science,135, pp. 538-546.

Molinero-Guerra, A., Cui, Y. J., Mokni, N., Delage, P., Bornert, M., Aimedieu, P., Tang, A. M., Bernier, F. and (2018). Investigation of the hydro-mechanical behaviour of a pellet/powder MX80 betonite mixture using an infiltration column. Engineering Geology, 243, pp. 18-25.

Pintado, X., Lloret, A., Romero, E., (2009). Assessment of the use of the vapour equilibrium technique in controlled-suction tests. Canadian Geotechnical Journal 46: 411-123

Saba, S., Cui, Y.J., Tang, A.M., and Barnichon, J.D., (2014). Investigation of the swelling behaviour of compacted bentonite–sand mixture by mock-up tests. Canadian Geotechnical Journal, 51(12), pp.1399–1412.

Sellin, P., and Leupin, O. X., (2013). The use of clay as an engineered barrier in radioactive-waste management – a review. Clays and Clay Minerals 61, No. 6, 477–498.

512 Schanz, T. and Al-Badran, Y., (2014). Swelling pressure characteristics of compacted Chinese
513 Gaomiaozi bentonite GMZ01. Soils and Foundations 54, No. 4, 748–759.

514 Villar, M.V., Rivas P. (1994). Hydraulic properties of montmorillonite-quartz and saponite-
515 quartz mixtures. Applied Clay Science 9, 1-9

516 Villar, M.V. (2005). MX-80 Bentonite. Thermo-Hydro-Mechanical Characterization
517 Performed at CIEMAT in the Context of Prototype Project. Ciemat technical report
518 1053. Madrid, Spain.

519 Villar, M. V., Gómez-Espina, R. and Guitiérrez-Nebot, L., (2012). Basal spacings of smectite
520 in compacted bentonite. Applied Clay Science 65–66, 95–105.

521 Wang, Q., Tang, A. M., Cui, Y. J., Delage, P., and Gatmiri, B., (2012). Experimental study on
522 the swelling behaviour of bentonite/claystone mixture. Eng. Geol., vol. 124, no. 1, pp.
523 59–66.

524 Wang, Q., Tang, A.M., Cui, Y.J., Barnichon, J.D. and Ye, W.M., (2013a). A comparative stud
525 on the hydro-mechanical behaviour of compacted bentonite/sand plug based on
526 laboratory and field infiltration tests. Eng. Geol. 162, 79–87.

527 Wang, Q., Tang, A.M., Cui, Y.J., Barnichon, J.D. Ye, W.M., (2013b). Investigation of the
528 hydro-mechanical behaviour of compacted bentonite/sand mixture based on the BExM
529 model. Computer and Geotechnics 54, 46-52.

530 Yahia-Aissa,M., Delage, P., Cui, Y.J., (2001). Suction–water relationship in swelling clays.
531 Clay Science for Engineering. In: Adachi, K., Fukue,M. (Eds.), Proceedings of the IS-
532 Shizuoka International Symposium on Suction, Swelling, Permeability and Structure of
533 Clays. Shizuoka, Japan Balkema, pp. 65–68.

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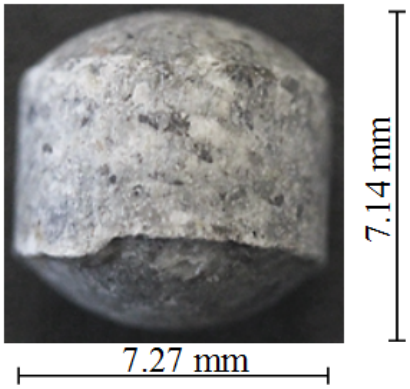

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567 Figure 1. Pellet at its initial state.

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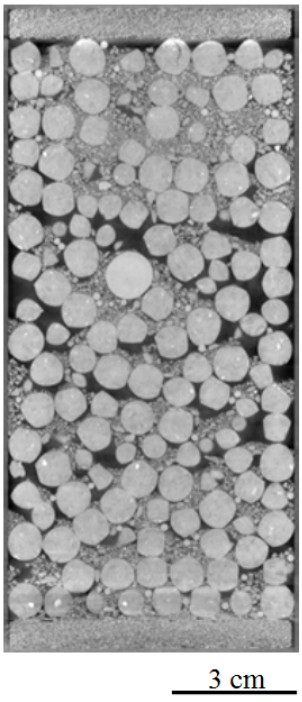

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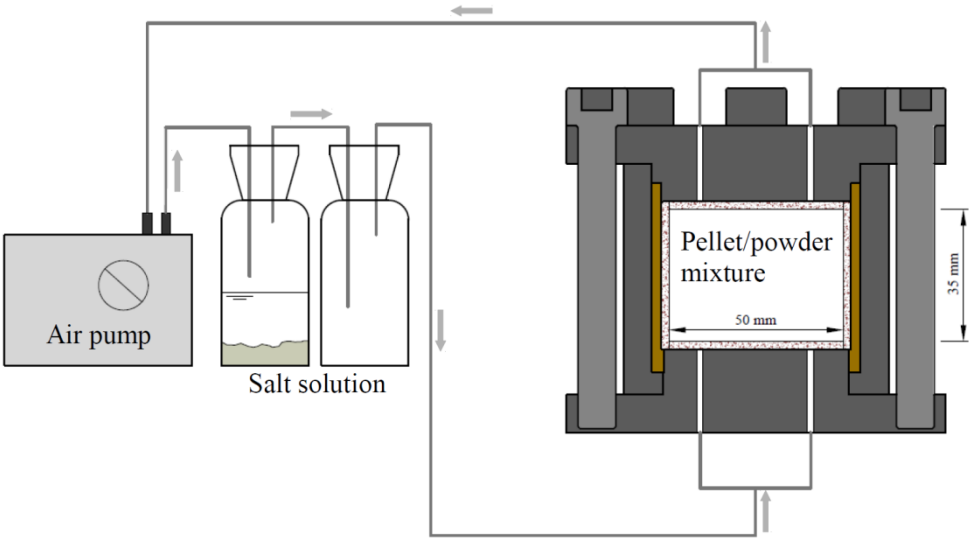
570 Figure 2. Vertical slice on the investigated pellet/powder bentonite mixture obtained by µ-CT
571 observations - Resolution: 50 µm/voxel.



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573 Figure 3. Schematic view of the special cell designed for determining the water retention
574 curve under constant-volume conditions.

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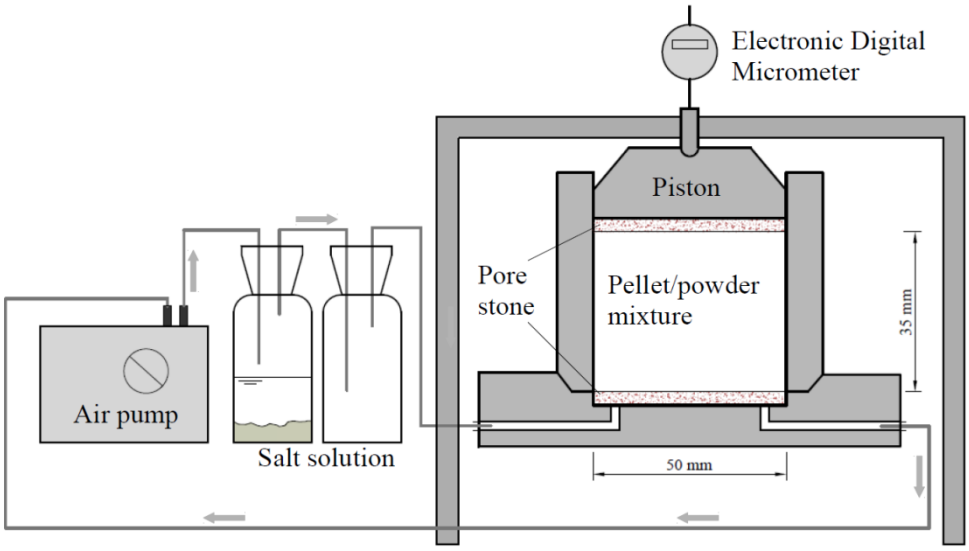

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577 Figure 4. Experimental setup for suction-controlled oedometer tests.

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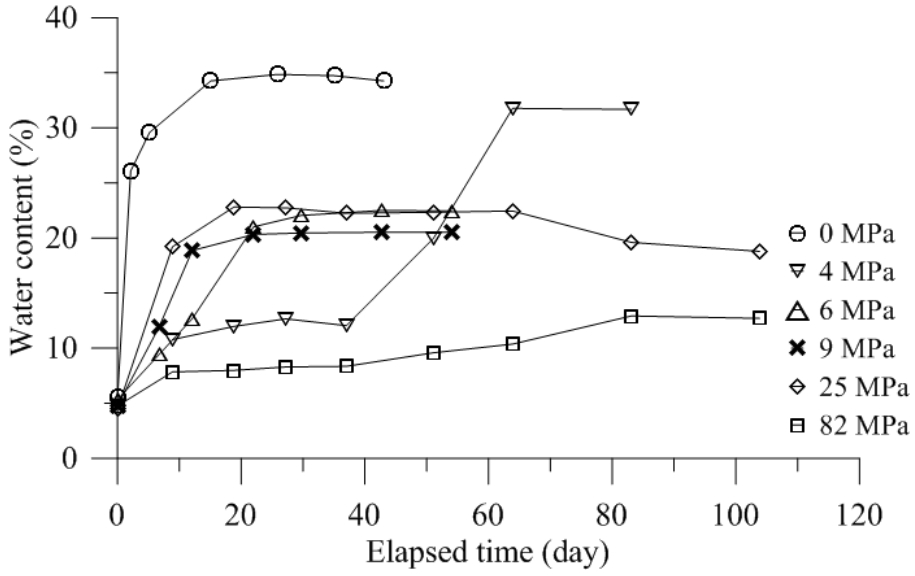

586

587 Figure 5. Evolution of water content with time for the pellet/powder bentonite mixture
588 submitted to different suctions under constant-volume conditions.

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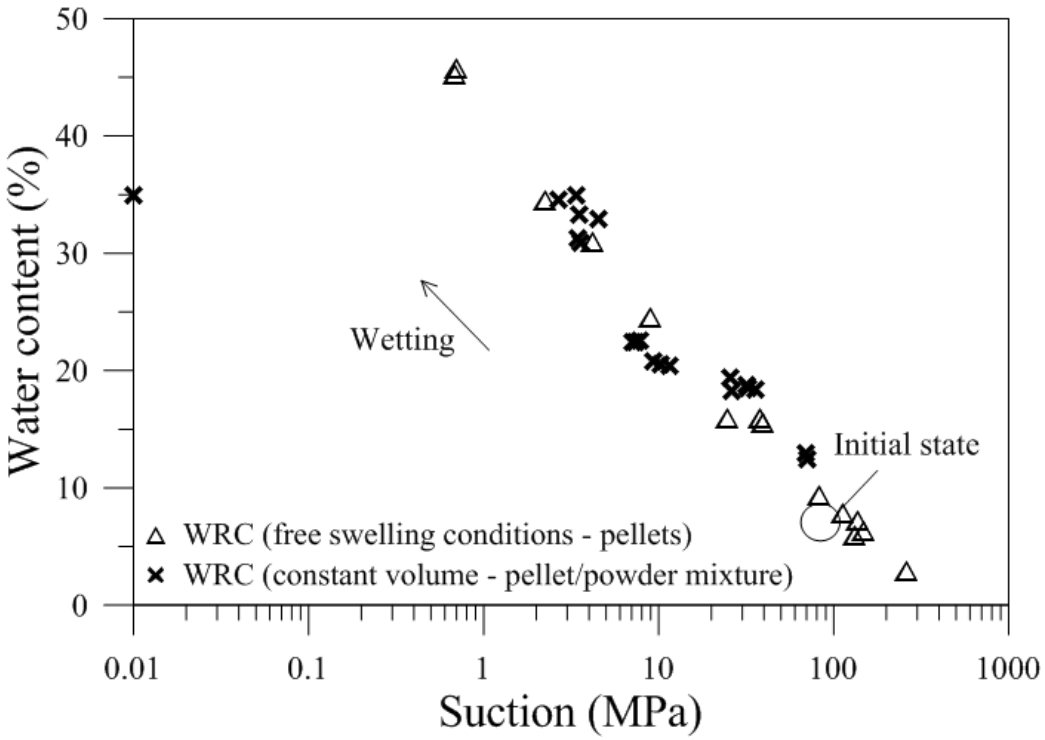

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591 Figure 6. Water retention properties under free swelling conditions and constant-volume
592 conditions.

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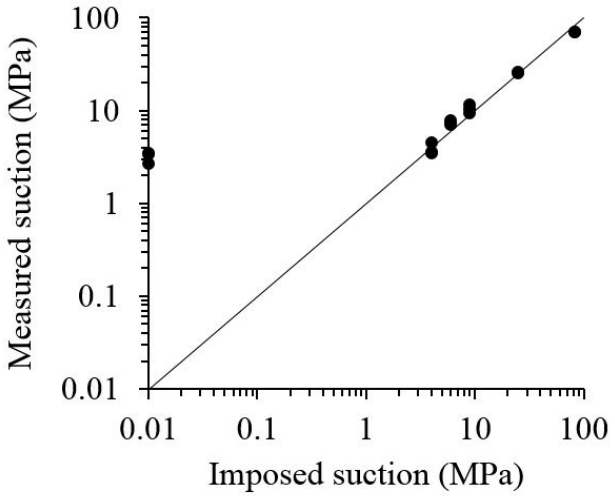

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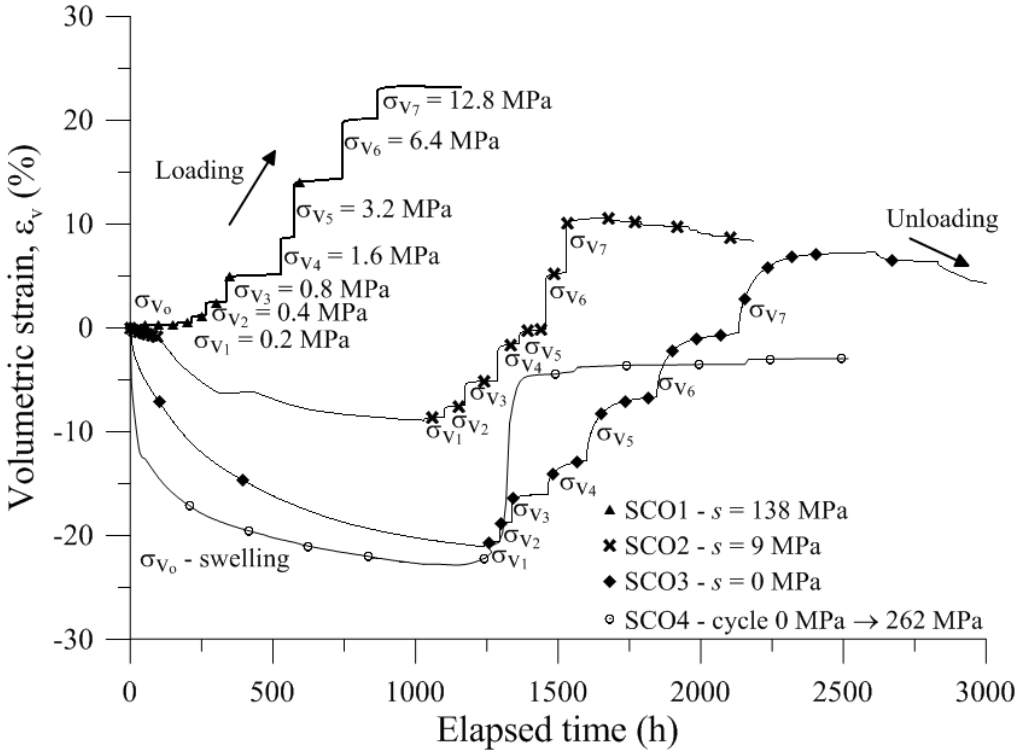

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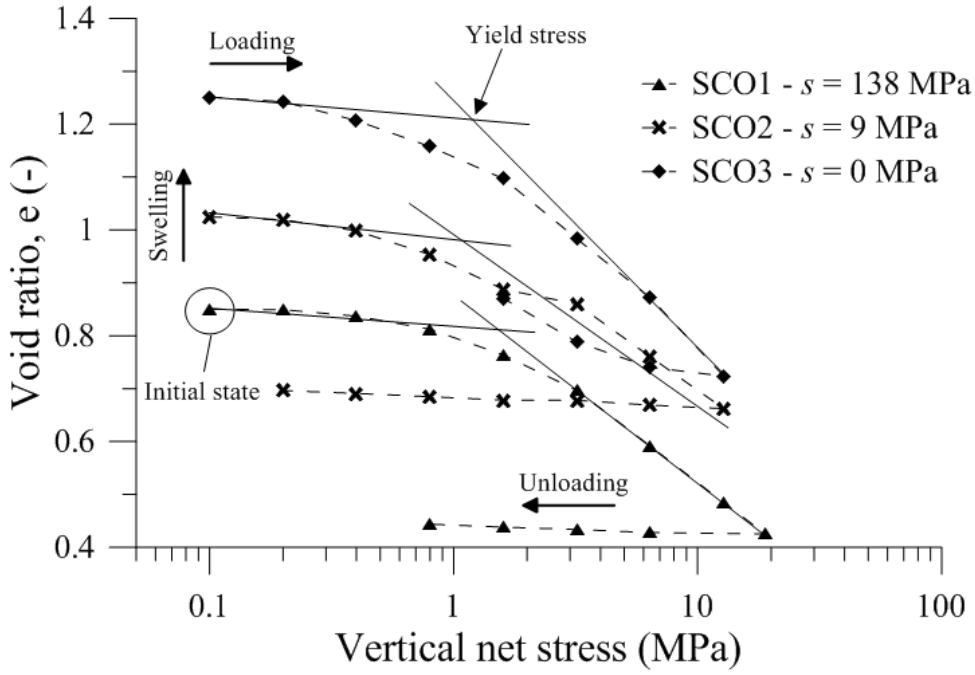

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603 Figure 9. Void ratio versus vertical stress for tests SCO1, SCO2 and SCO3.

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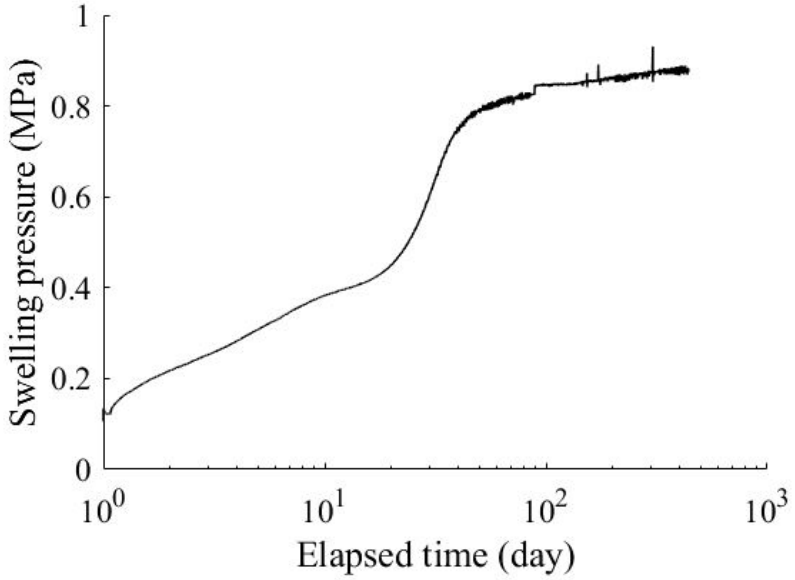

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606 Figure 10. Evolution of axial swelling pressure with time in test CV1.

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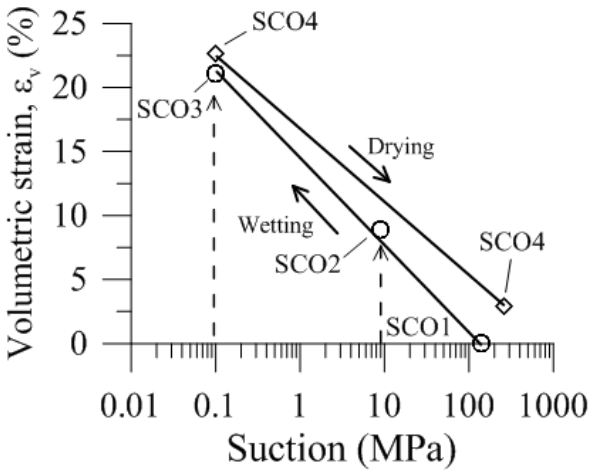

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609 Figure 11. Volumetric strains measured at different suctions after equilibrium at the imposed
610 suction for the different tests.

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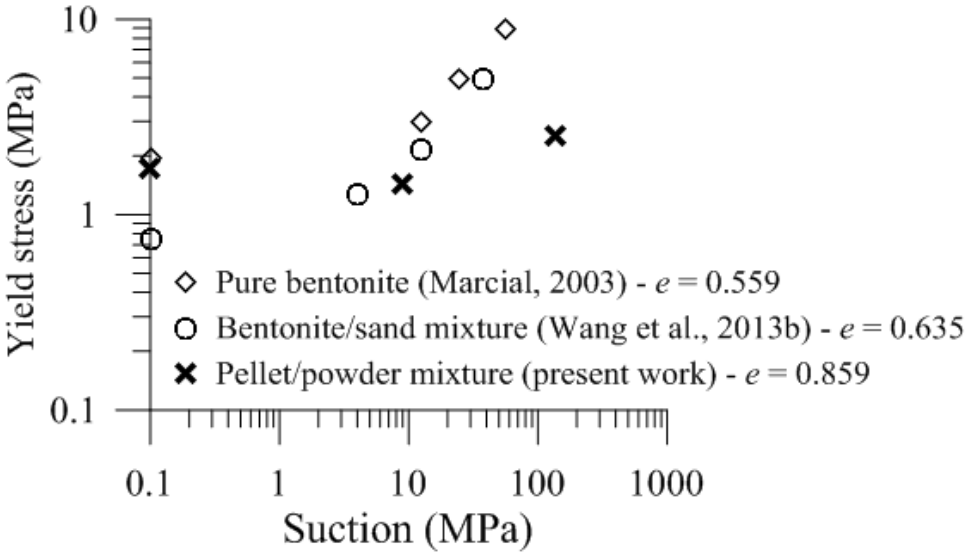

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613 Figure 12. Yield stress versus suction for compacted bentonite with initial void ratio of 0.559
614 (Marcial, 2003), compacted bentonite/sand mixture with initial void ratio of 0.635 (Wang et al.,
615 2013b) and pellet/powder mixture with initial void ratio of 0.859 (present work).

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622 Table 1. Test program for investigating the compressibility and swelling properties of
623 pellet/powder bentonite mixture.

Test No.	Hydration condition	Suction (MPa)	Investigated property	Method
SCO1	Constant water content	138	Compressibility	-
SCO2	Vapor transfer	9	Compressibility	-
SCO3	Injected deionized water	0	Compressibility and swelling	Swell-consolidation
SCO4	Injected deionized water and vapor transfer	0 → 262	Swelling	-
CV1	Injected synthetic water	0	Swelling	Constant-volume

624

625 Table 2. Receipt for preparing the synthetic water.

Components	$NaHCO_3$	Na_2SO_4	NaCl	KCl	$CaCl_2$ $2H_2O$	$MgCl_2O6H_2O$	$SrCl_26H_2O$
Mass (g) per litre of solution	0.28	2.216	0.615	0.075	1.082	1.356	0.053

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