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Abstract: This paper investigates the swelling mechanism of Callovo-Oxfordian (COx) claystone grain in its mixtures with bentonite which are potential candidate for engineered barriers in the French high-level radioactive waste disposal. The swelling pressures of MX80 bentonite/COx claystone mixtures with various bentonite fractions and dry densities were first experimentally determined by carrying out infiltration tests under constant-volume conditions. According to the experimental results, the claystone void ratio after hydration in the mixtures was estimated by taking into account the bentonite–claystone grain interaction. Then, the volume change of claystone grain under different pressures applied by swollen bentonite was determined. The volumetric strain of claystone grain was found to decrease linearly with the increase of pressure. This allowed the indirect determination of the swelling pressure of claystone grain. The determined value was found to be comparable to that estimated from the relationship between the void ratio and the swelling pressure in the direction perpendicular to the stratification of intact COx claystone, confirming the identified swelling mechanism of bentonite/claystone mixtures.

Keywords: bentonite/claystone mixture; claystone void ratio; volumetric strain; swelling pressure of claystone grain

List of symbols

B bentonite fraction

e void ratio

e_c claystone void ratio

e_c^0 initial void ratio of claystone grain

G_{sc} specific gravity of claystone

K_0 earth pressure coefficient at rest

OCR over-consolidation ratio

P_s measured global swelling pressure of mixture

$P_{s\text{-}mean}$ mean swelling pressure of claystone

$P_{s\text{-}per}$ swelling pressure in the direction perpendicular to the stratification of intact claystone

ρ_w water unit mass

ρ_{db} bentonite dry density

ρ_{dc} claystone dry density

ρ_{dc}^0 initial claystone dry density

ρ_{dm} mixture dry density

ε_v claystone volumetric strain of claystone

φ_c effective internal friction angle

1 Introduction

The French Radioactive Waste Management Agency (Andra) is currently envisaging to construct a geological repository for high-level radioactive waste in the Callovo-Oxfordian (COx) claystone formation [1]. The disposal galleries are planned to be excavated at a depth of 490 m in the 150-m-thick layer of COx claystone. To seal or backfill these galleries after the emplacement of waste canisters, pure bentonite or bentonite/sand mixture may be employed. To reduce the excavation wastes and to have lower costs, excavated COx claystone has been proposed to partially or completely replace the commercial bentonite as sealing/backfilling material. Once the galleries are closed, the crushed claystone or its mixture with bentonite is expected to exhibit a swelling pressure high enough to stabilize the surrounding rock mass [7, 13, 18].

Over the last few decades, the swelling pressure of pure bentonite and bentonite/sand mixtures were widely studied [3-5, 9]. It was found that the swelling pressure increased linearly with the increase of dry density in a semi-logarithmic plane. Additionally, for each specific bentonite, the relationship between the swelling pressure and the final bentonite dry density could be described by a unique relationship, regardless of sand fraction [2, 11]. The swelling pressure of bentonite/sand mixtures was principally dominated by the matrix of bentonite, while the sand grains remained inert without any contribution to the global swelling pressure. Unlike the inert sand, the COx claystone contains a significant amount of clay minerals. Upon contact with water, both the bentonite and claystone grains adsorbed water and the interaction between them could occur. Zeng et al. [13] argued that the swelling of claystone was conditioned by the swollen bentonite due to the much higher swelling capacity of bentonite. However, this swelling mechanism of claystone grain in its mixtures

77 with bentonite has not been analysed in-depth.

78 In this paper, a series of constant-volume swelling pressure tests were performed on the MX80
79 bentonite/COx claystone mixtures with various bentonite fractions and dry densities. By considering
80 the interaction between bentonite and claystone in the development of global swelling pressure, the
81 claystone void ratio in the mixture after hydration was calculated and the relationship between the
82 claystone grain volume change and the pressure applied by swollen bentonite was determined.
83 Subsequently, the swelling pressure of crushed claystone grains was indirectly determined and then
84 compared with that estimated from the measured swelling pressure in the direction perpendicular to
85 the stratification of intact COx claystone.

86 **2 Materials and methods**

87 The mixtures of MX80 bentonite and COx claystone powders were used in this study. The MX80
88 bentonite was taken from Wyoming, USA. Prior to the experiment, the bentonite was crushed to
89 powders with a maximum grain size of 2.0 mm. The COx claystone was extracted from the Bure
90 Underground Research Laboratory. The subsequent processing of excavated COx claystone
91 comprised the crushing and sieving passing through 2.0 mm sieve. The grain size of processed COx
92 claystone powders used in this work is in the range of 0.08-2.0 mm. The grain size distributions of
93 the bentonite and claystone determined by dry-sieving method are shown in Fig. 1. The mean particle
94 sizes D_{50} of the bentonite and claystone powders are 0.55 and 0.58 mm, respectively. The physico-
95 chemical properties of the tested materials are listed in Table 1. The initial suctions of the bentonite
96 and claystone powders were also measured by hygrometer WP4, equal to 101 and 27 MPa,
97 respectively. Using a pycnometer with a non-aromatic hydrocarbon fluid (Kerdane), the apparent

98 volume of soil grains at a given dry mass was measured and the respective average dry densities of
99 bentonite and claystone grains were thus determined, equal to 2.00 and 2.31 Mg/m^3.

100 **Table 1** Physico-chemical properties of MX80 bentonite and COx claystone

Soil property	MX80 bentonite	COx claystone
Water content (%)	11.4	6.1
Specific gravity	2.76	2.70
Liquid limit (%)	494	41
Plastic limit (%)	46	24
Clay-size fraction (%)	86	26
Main clay minerals	Smectite	Interstratified illite-smectite

101 The bentonite and claystone powders at various proportions of 70/30, 50/50, 30/70, 20/80, 10/90
102 in dry mass were first mixed thoroughly. As shown in Table 2, the corresponding bentonite fractions
103 (B) of the mixtures were 70, 50, 30, 20 and 10%. Subsequently, the mixtures and pure claystone were
104 statically compacted at a control rate of 0.05 mm/min to reach various target dry densities. The
105 diameter and height of the compacted samples were 50 and 10 mm, respectively. After compaction,
106 the samples were transferred to the stainless steel cell (50 mm in diameter) and the two sides of
107 samples were covered by two filter papers and two porous discs (Fig. 2). At the top, the samples were
108 restrained by a piston blocked using screws. Synthetic site water was injected from the bottom of the
109 cell at constant water head of about 1 m (the corresponding water pressure was about 10 kPa). The
110 synthetic site water has the same chemical composition as the site water in Bure and it was prepared
111 by mixing the relevant chemical components (Table 3) with de-ionized water until full dissolution.
112 The samples were hydrated under constant-volume conditions and the expelled air/water flowed
113 through the air/water outlets at the top. During the hydration, the vertical force was monitored using
114 a force transducer (accuracy ±0.01 kN) installed under the cell. Note that the maximum vertical

115 displacement of the apparatus was quite small, estimated at 0.0049 mm. Thus, the volume change of

116 samples due to the deformation of apparatus was ignored.

117 **Table 2** Test program and main results

Test No.	Bentonite fraction B (%)	Dry density of sample ρ_{dm} (Mg/m^3)	Initial water content w_m (%)	Claystone void ratio e_c	Volumetric strain of claystone grain ε_v (%)	Final swelling pressure P_s (MPa)
S01	70	1.38	9.8	0.08	-7.8	0.43
S02	70	1.50	9.8	0.13	-2.9	1.11
S03	70	1.63	9.8	0.07	-8.1	2.53
S04	70	1.71	9.8	0.02	-12.4	3.94
S05	50	1.27	8.8	0.49	27.4	0.13
S06	50	1.56	8.8	0.30	11.5	0.85
S07	50	1.73	8.8	0.18	1.1	2.10
S08	50	1.76	8.8	0.19	2.2	2.91
S09	30	1.50	7.7	0.18	26.2	0.22
S10	30	1.60	7.7	0.47	20.6	0.46
S11	30	1.68	7.7	0.41	16.4	0.78
S12	30	1.79	7.7	0.36	10.8	1.59
S13	30	1.89	7.7	0.30	5.0	2.72
S14	30	1.99	7.7	0.23	0.9	5.23
S15	20	1.60	7.2	0.18	27.3	0.26
S16	20	1.69	7.2	0.49	20.5	0.44
S17	20	1.77	7.2	0.41	15.8	0.70
S18	20	1.88	7.2	0.35	11.8	1.93
S19	10	1.61	6.6	0.31	33.7	0.14
S20	10	1.68	6.6	0.56	28.7	0.23
S21	10	1.78	6.6	0.50	21.5	0.39
S22	10	1.90	6.6	0.42	15.5	1.07
S23	0	1.80	6.1	0.35	28.3	0.15
S24	0	1.90	6.1	0.50	21.8	0.47
S25	0	1.99	6.1	0.42	16.3	0.79

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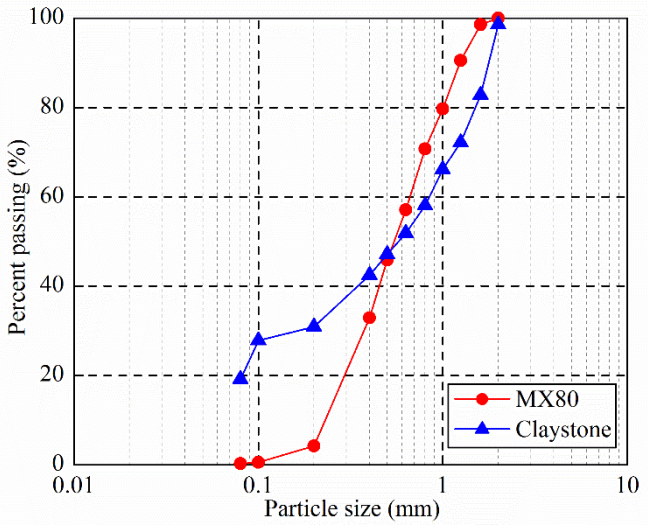

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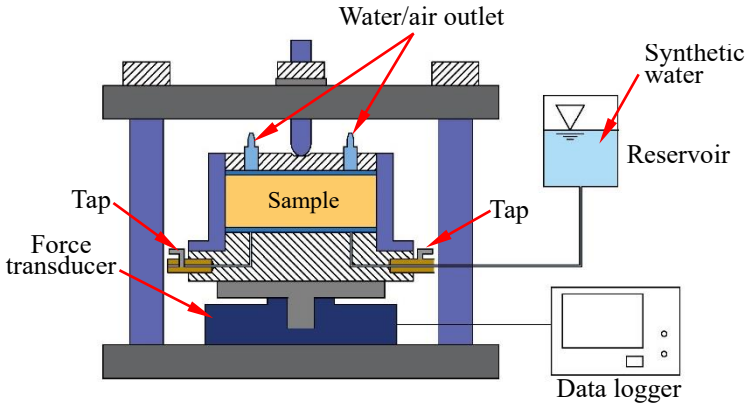
120 **Fig. 1** Grain size distributions of MX80 bentonite and COx claystone



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122 **Fig. 2** Layout of the experimental setup for swelling pressure tests

123 **Table 3** Chemical composition of the synthetic site water

Component	NaCl	$NaHCO_3$	KCl	$CaSO_4 \cdot 2H_2O$	$MgSO_4 \cdot 7H_2O$	$CaCl_2 \cdot 2H_2O$	Na_2SO_4
Content (g/L)	1.950	0.130	0.035	0.630	1.020	0.080	0.700

124 **3 Experimental results and discussion**

125 *3.1 Global swelling pressure*

126 Fig. 3 depicts the typical evolution of the global swelling pressure of samples with various bentonite

127 fractions and dry densities. On the whole, the global swelling pressure of the samples with high

128 bentonite fractions (larger than 50%) and high dry densities (larger than 1.50 Mg/m^3) increased with

129 elapsed time and then tended to stabilise. By contrast, the swelling pressure of the samples with low

130 bentonite fractions (smaller than 20%) and low dry densities (smaller than 1.77 Mg/m^3) increased

quickly at the beginning and reached a peak. Then, it decreased and tended to stabilise.

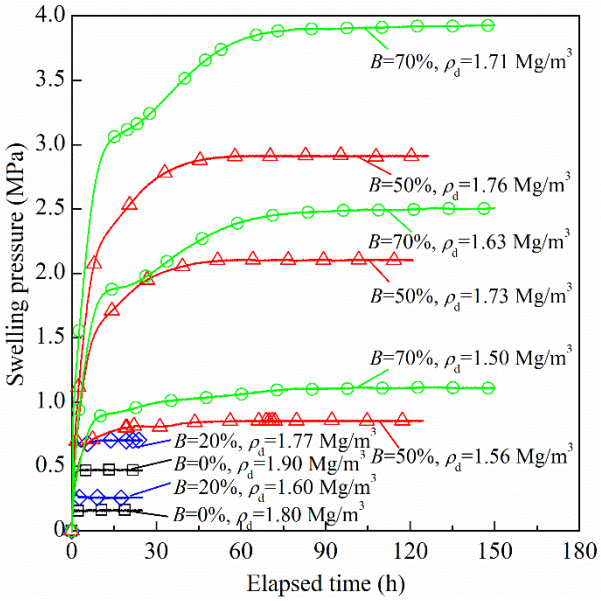
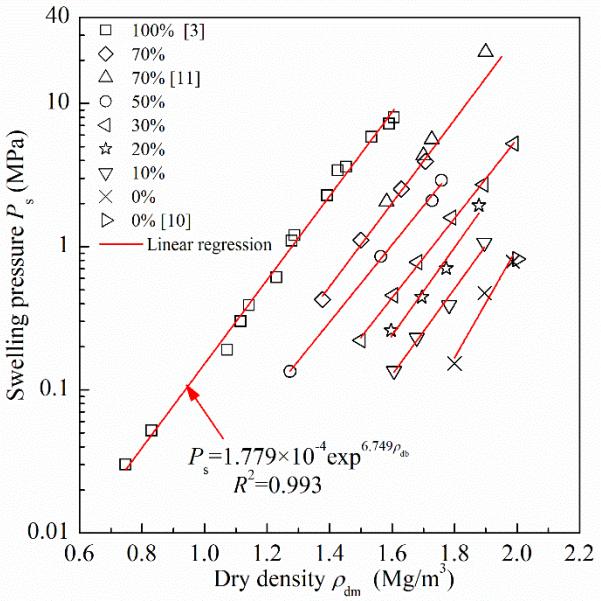


Fig. 3 Typical evolutions of the global swelling pressure of samples with various bentonite fractions and dry densities

The swelling pressure tests ended after 25-150 h and the samples became saturated according the results of Zeng et al. [14]. The variation of final swelling pressure with respect to the dry density of samples is presented in Fig. 4 and compared with the results from other researchers [3, 10, 11]. It is observed that the final swelling pressure of samples increased linearly with the increase of dry density in a semi-logarithmic plane. At the same dry density, the swelling pressure of pure bentonite was 100 times larger than that of pure claystone. For the bentonite/claystone mixtures, the higher the bentonite fraction, the larger the final swelling pressure.



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143 **Fig. 4** Variation of the final swelling pressure with sample dry density

144 *3.2 Volume change behaviour of claystone grains*

145 When the bentonite/claystone mixtures were hydrated, the bentonite grains are expected to rapidly
146 swell and fill up the voids in the mixtures, thanks to their extremely high swelling capacity, while the
147 claystone grains are expected to adsorb water under the pressure applied by the swollen bentonite [13,
148 15]. This implies that the pressure at the interface of bentonite and claystone was governed by the
149 bentonite property. Note that the water pressure was quite low and the pressure at the interface of
150 bentonite and claystone could be considered as an effective stress. To clarify the role of the claystone
151 in the mixture, an additional assumption was made: the pressure at the interface of bentonite and
152 claystone was equal to the global swelling pressure (Fig. 5a and b), as suggested by Yang et al. [12]
153 when dealing with the consolidation behaviour of a lumpy granular soil under one-dimensional
154 condition. Thereby, given the measured global swelling pressure of mixture (P_s in MPa), the final
155 bentonite dry density in the mixture (ρ_{db} in Mg/m^3) can be estimated according to the relationship
156 between the swelling pressure and the final dry density of pure bentonite (Fig. 4):

$$P_s=1.779\times10^{-4}\exp^{6.749\rho_{db}} \quad (1)$$

Based on the binary composition of the mixture, the final claystone dry density (ρ_{dc} in Mg/m^3) in the mixture can be deduced using Eq. (2):

$$\rho_{dc}=\frac{\rho_{db}\rho_{dm}(1-B/100)}{\rho_{db}-\rho_{dm}(B/100)} \quad (2)$$

where ρ_{dm} is the final mixture dry density (expressed in Mg/m^3) and ρ_{db} is the final bentonite dry density (expressed in Mg/m^3).

For pure claystone (B=0), the COx claystone swelled and filled up the voids between grains, becoming continuous (Fig. 5c). Thus, the final COx claystone dry density after saturation is equal to the global dry density.

Correspondingly, the final claystone void ratio e_c in the mixture and pure claystone can be determined:

$$e_c=\frac{G_{sc}\rho_w}{\rho_{dc}}-1 \quad (3)$$

where G_{sc} is the specific gravity of claystone and ρ_w is the water unit mass.

In order to describe the volume change behaviour of claystone grain after hydration, the claystone volumetric strain (ε_v) was determined using Eq. (4):

$$\varepsilon_v=\frac{e_c-e_c^0}{(1+e_c^0)}=\frac{\rho_{dc}^0}{\rho_{dc}}-1 \quad (4)$$

where e_c^0 is the initial void ratio of claystone grain and ρ_{dc}^0 is the initial dry density (equal to 2.31 Mg/m^3). According to Eqs. (1), (2), (3) and (4), the claystone void ratios and the volumetric strains of claystone for all the samples with various bentonite fractions and dry densities were calculated and the calculated results are summarized in Table 2. For further analysis, the results of Wang et al. [11] were collected and analysed in the same fashion. Fig. 6 depicts the variation of volumetric strain with

the pressure applied by the swollen bentonite. It appears that the volumetric strain of claystone decreased with the increase of pressure, regardless of the bentonite fraction. Moreover, the claystone grains swelled under low pressures and collapsed under high pressures.

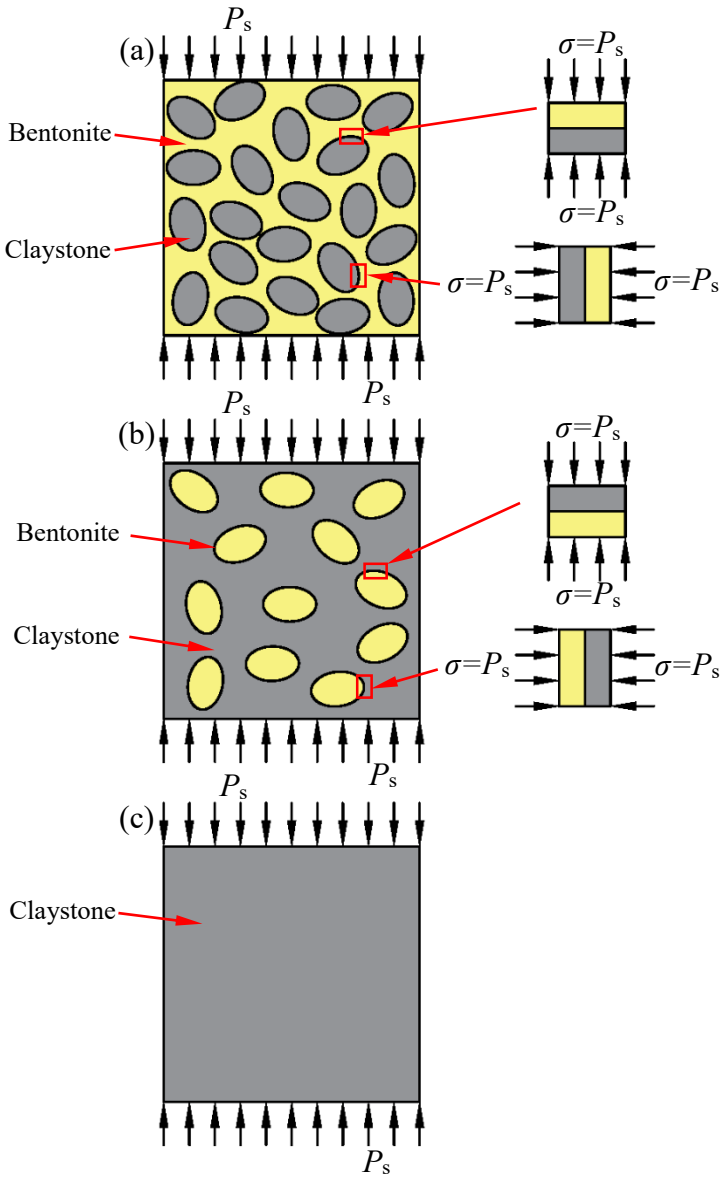

Fig. 5 Structure sketch of the samples after saturation: (a) mixtures with a large bentonite fraction, (b) mixtures with a small bentonite fraction, and (c) pure claystone. Note: the zones for bentonite and claystone after full saturation include voids in them

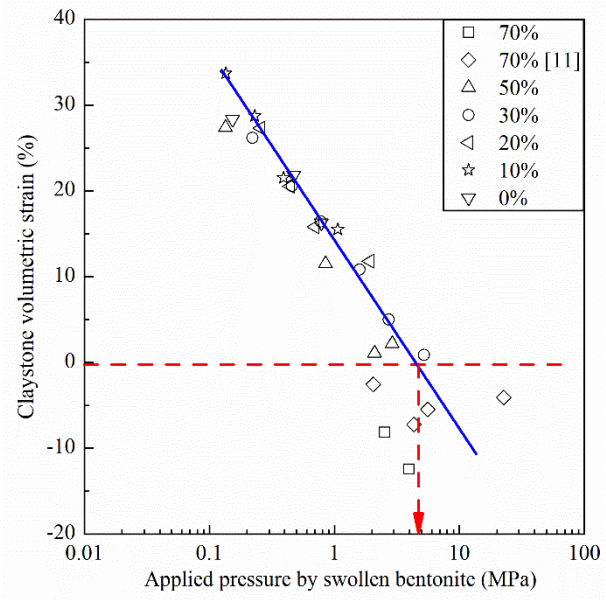

Fig. 6 Relationship between the claystone volumetric strain and the applied pressure by swollen bentonite

3.3 Swelling pressure of claystone grains

To further verify the swelling mechanism of claystone in its mixtures with bentonite, the swelling pressure of claystone grain was indirectly determined and then compared with that measured on intact COx claystone. According to the load-swell method, the volume changes of claystone grains subjected to various stresses were measured. This allowed the indirect determination of the swelling pressure of claystone grains. The swelling pressure corresponds to the stress at the intersection of the volumetric strain curve (represented by the blue solid line in Fig. 6) with the no volume change line (represented by the dash line in Fig. 6). A swelling pressure of about 4.9 MPa was thus determined for the claystone grain. Note that this swelling pressure referred to a mean value of those in all directions.

Using constant-volume, load-swell and Cui' methods, Zhang et al. [19] performed a series of swelling pressure tests on intact COx claystone, which had a similar water content to the crushed claystone powders used in this study (6.1%). They found that there was a good relationship between

the swelling pressure ($P_{s\text{-}per}$ in MPa) and void ratio (e) (Fig. 7):

$$P_{s\text{-}per}=1.365\times10^{6}\exp^{-71.562e} \quad (5)$$

According to Eq. (5), the intact COx claystone with a void ratio of 0.17 corresponded to a swelling pressure of 7.11 MPa. It is worth noting that this swelling pressure was measured in the direction perpendicular to the stratification of claystone and did not represent the mean value. The measured swelling pressure corresponded to an effective stress because the pore water pressure could be ignored during the testing [17]. Moreover, in the swelling pressure test, the intact claystone was placed in oedometer cell and it swelled under zero lateral deformation condition or K_0 condition. According to the empirical relationship proposed by Mayne & Kulhawy [6], the earth pressure coefficient at rest K_0 (ratio of horizontal and vertical effective stresses) for over-consolidated soil can be estimated using Eq. (6):

$$K_0=(1-\sin\varphi_c)\mathrm{OCR}^{\sin\varphi_c} \quad (6)$$

where OCR and φ_c are the over-consolidation ratio and the effective internal friction angle, respectively. The over-consolidation ratio is defined as the ratio of pre-consolidation stress to the current vertical stress. Based on the results from oedometer compression test on intact COx claystone, in which the loading direction was perpendicular to the stratification of claystone [8], the pre-consolidation stress was estimated at 8.5 MPa and the OCR value at 1.20 under a swelling pressure of 7.11 MPa. Zhang et al. [16] investigated the shear strength of intact COx claystone by carrying out triaxial compression tests and found that the internal friction angle of intact COx claystone was about 24°. From the over-consolidation ratio and the effective internal friction angle, the K_0 value was estimated at 0.64. Substituting the swelling pressure in the direction perpendicular to the stratification

and K_0 value into Eq. (7) yields the mean swelling pressure ($P_{s\text{-mean}}$ in MPa):

$$P_{s\text{-mean}}=P_{s\text{-per}}\frac{1+2K_0}{3} \quad (7)$$

The estimated mean swelling pressure of intact COx claystone was 5.39 MPa, which was quite close to the value of 4.9 MPa indirectly determined from the swelling pressure tests on the bentonite/claystone mixtures. This confirmed the reliability of the assumptions made above and the relevance of the identified swelling mechanism for claystone in its mixtures with bentonite. Upon hydration, the bentonite grains in the mixtures would swell rapidly due to their high swelling capacity while the claystone grains adsorbed water and then swelled or collapsed under the pressure applied by the previously swollen bentonite. The identified mechanism can be extended to other binary mixtures composed of two clays with extremely different swelling capacities.

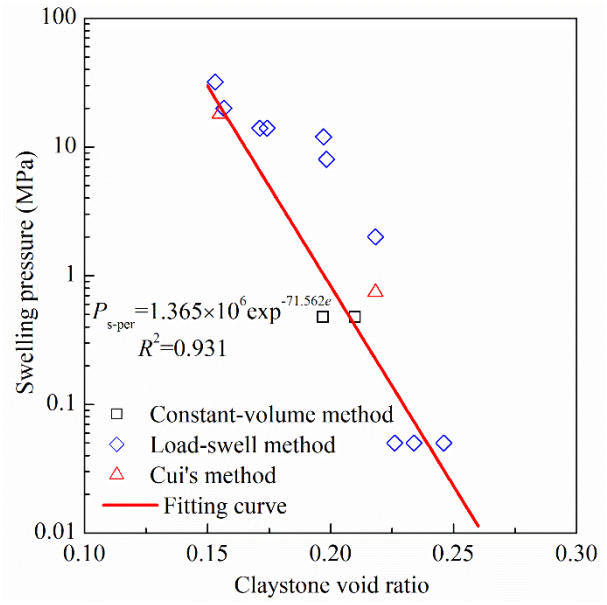

Fig. 7 Swelling pressure versus claystone void ratio of intact claystone (modified after Zhang et al. [19])

4 Conclusions

The swelling pressures of MX80 bentonite/COx claystone mixtures with various bentonite fractions and dry densities were experimentally investigated in the laboratory. By considering the interaction

between bentonite and claystone grains during hydration, the claystone void ratio in the mixtures after hydration was estimated based on the relationship between the swelling pressure and dry density of pure bentonite. Comparison of the claystone void ratio after hydration with the initial one allowed the volumetric strains of claystone grains under different stresses to be obtained. It was found that the volumetric strain decreased linearly with the increase of pressure applied by the swollen bentonite. According to this relationship, the mean swelling pressure of claystone grain was indirectly determined.

Additionally, the mean swelling pressure of claystone grain was estimated according to the relationship between the void ratio and the swelling pressure in the direction perpendicular to the stratification of intact COx claystone. The estimated value was found to be comparable to that indirectly determined from the swelling pressure tests on bentonite/claystone mixtures. This confirmed the identified swelling mechanism of COx claystone grain in its mixtures with bentonite: the claystone grains adsorbed water under the pressure applied by the previously swollen bentonite.

Notably, the swelling pressure of claystone with a specified water content was determined in this study. The influence of initial water content on the swelling pressure of claystone powders will be investigated in further studies.

Acknowledgments

The authors thank the Chinese Scholar Council (CSC) and the French Radioactive Waste Management Agency (Andra) for the financial supports.

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