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1 **Investigation of swelling pressure of bentonite/claystone mixture in the full range**
2 **of bentonite fraction**

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21 **Abstract:** MX80 bentonite/Callovo-Oxfordian (COx) claystone mixture has been proposed as a
22 sealing/backfilling material in a deep geological repository of radioactive waste in France. A
23 good understanding of the swelling behaviour of this mixture is essential when evaluating the
24 long-term performance of the repository. In this work, the swelling pressure of MX80
25 bentonite/COx claystone mixture was investigated by the constant-volume method for a full
26 range of bentonite fraction. Results show that the swelling of claystone in the mixture can be
27 inhibited by bentonite and its contribution to the global swelling pressure depends on the
28 bentonite fraction. For the mixture with more than 70% bentonite, claystone behaves as an inert
29 material, and its contribution to the global swelling pressure can be ignored. However, for the
30 mixture with less bentonite, the swelling of claystone will significantly contribute to the global
31 swelling pressure. A method was proposed allowing the swelling pressure of bentonite/claystone
32 mixture to be predicted in the full range of bentonite fraction.

33

34 **Keywords:** bentonite/claystone mixture; swelling pressure; bentonite dry density; inhibition
35 effect; prediction method

36 **1 Introduction**

37 Deep geological disposal has been adopted for radioactive waste in many countries such as China,
38 Belgium, France, Germany, Japan, Sweden, etc. (Gray et al., 1984; Dixon et al., 1985; Sellin and
39 Leupin, 2013). To ensure the isolation of radioactive waste from the surrounding environment,
40 bentonite-based materials are often considered as possible sealing/backfilling materials because of
41 their favorable swelling characteristics and low permeability (Pusch, 1982; Dixon et al., 1985;
42 Komine and Ogata, 1994, 1999; Villar and Lloret, 2008). Once the disposal galleries are closed, pore
43 water from the host rock will progressively infiltrate into the sealing/backfilling materials. They will
44 swell and seal the technological voids between the blocks of compacted bentonite-based materials or
45 between the blocks and the canisters/the host rock (Bian et al., 2018). Afterwards, swelling pressure
46 will develop (Pusch, 1982; Wang et al., 2012). This swelling pressure must be high enough to ensure
47 the good sealing performance, but lower than the in situ minor stress in the host rock (Saba et al.,
48 2014). Therefore, the swelling pressure of bentonite-based materials becomes a key factor in the
49 design of deep geological repositories.

50 In the past decades, pure bentonite or bentonite/sand mixture was widely investigated as a
51 sealing/backfilling material for the easy control of its swelling pressure (Komine and Ogata, 1994,
52 1999). A number of studies have been conducted on the swelling pressure of different compacted
53 bentonites and their mixture with sand, such as Kunigel-V1 bentonite (Sun et al., 2009), MX80
54 bentonite (Karnland et al., 2008; Saba et al., 2014), Calcigel bentonite (Agus and Schanz, 2008),
55 Tsukinuno bentonite (Komine and Ogata, 1999) and GMZ bentonite (Ye et al., 2007; Cui et al., 2012;
56 Sun et al., 2015, 2017). The swelling pressure of bentonite/sand mixture has been characterized
57 quantitatively based on the montmorillonite void ratio (Sun et al., 2009; Sun et al., 2015, 2017), dry

58 density of bentonite (Dixon et al., 1985; Lee et al., 1999; Agus and Schanz, 2008), effective dry
59 density of montmorillonite (Dixon et al., 2002; Powell et al., 2013), and initial degree of saturation of
60 montmorillonite (Rao and Ravi, 2015).

61 The French National Agency for Nuclear Waste Management (ANDRA) proposed to use a
62 mixture of bentonite and excavated Callovo-Oxfordian (COx) claystone as the sealing/backfilling
63 material. This aims to reduce the excavation waste and to better ensure the compatibility of
64 chemistry with the host rock (Tang et al., 2011; Wang et al., 2012, 2014). Wang et al. (2012) worked
65 on a MX80 bentonite/crushed COx claystone mixture at a proportion of 70/30 in dry mass and found
66 that the unique relationship between the swelling pressure and bentonite dry density could be
67 extended to this mixture, the contribution of claystone to swelling pressure being negligible.
68 However, the swelling mechanism of bentonite/claystone mixtures with bentonite fractions lower
69 than 70% has not been well understood.

70 In this study, the swelling pressure of MX80 bentonite/crushed COx claystone mixture with
71 different dry densities and bentonite fractions was studied. The role of claystone in the development
72 of swelling pressure of the mixture was analyzed, allowing an inhibition factor to be defined for
73 describing the effect of bentonite on the swelling of claystone. A novel method was then proposed to
74 predict the swelling pressure of bentonite/claystone mixture in the full range of bentonite fraction.

75 **2 Materials and experimental methods**

76 *2.1 Materials*

77 The commercial MX80 bentonite tested in this study was extracted from Wyoming, USA. Table 1
78 summarizes the basic physical and chemical properties of MX80 bentonite. The maximum grain size
79 of the bentonite powder is 2 mm and the clay-size fraction ($< 2 \mu\text{m}$) represents 84%. The density of

80 bentonite grains was measured to be 2.00 Mg/m^3 by immersing clay powders into a non-aromatic
81 hydrocarbon fluid (Kerdane) using a pycnometer. The initial suction measured by hygrometer WP4
82 was 101 MPa.

83 The COx claystone was extracted at around 490 m depth from the Underground Research
84 Laboratory (URL) at Bure, France. The claystone contains 40-45% clay minerals (mainly
85 interstratified illite-smectite), 30% carbonates and 2-30% quartz and feldspar, with a specific gravity
86 of 2.70 (Fouché et al., 2004). It was crushed into fine powders with grain size less than 2.0 mm.
87 More than 30% grains were smaller than $2 \mu\text{m}$. The density of the claystone grains is 2.31 Mg/m^3 .
88 The initial suction measured by hygrometer WP4 was 27 MPa.

89 In this study, a synthetic water (see Table 2 for the receipt of preparation), which has the same
90 chemical composition as the site groundwater from the URL in Bure, was used for hydration. The
91 total mass of dissolved solids is 4.545 g/L, which corresponds to a salinity (ratio of salt mass to
92 solution mass) lower than 0.5%.

93 *2.2 Sample preparation*

94 A series of tests were conducted on compacted claystone and MX80 bentonite/claystone mixtures
95 with bentonite fractions of 10, 20, 30, 50, and 70% in dry mass. The bentonite and claystone, with
96 initial water contents of 11.4% and 6.1%, respectively, were mixed for more than 10 min to reach a
97 homogeneous state. Subsequently, samples were statically compacted in a cylindrical mold at a
98 constant displacement rate of 0.05 mm/min to reach the target dry density (Table 3). This rate was
99 chosen to ensure zero air over pressure in soil during compaction. After decompression, the samples
100 had a diameter of 50 mm and a height of 10 mm.

101 *2.3 Experimental methods*

102 Twenty-five swelling pressure tests were carried out using a constant-volume cell at a temperature of
103 $20\pm 1^\circ\text{C}$. The details about the constant-volume cell can be found in Saba et al. (2014). The
104 compacted samples were pushed into the testing cell from the mold and placed between two porous
105 stones and filter papers. The top cap was locked by a screw. The samples were hydrated from bottom
106 through the water inlet connected to the synthetic water reservoir. The swelling pressure was
107 monitored by a force transducer mounted below the test cell and all data were collected by a data
108 logger. The maximum deformations of the apparatus and filter papers in this study were estimated to
109 be 0.0049 and 0.0169 mm, corresponding to a small reduction of 2.3% in maximum swelling
110 pressure. Thus, the volume change of samples throughout the water-uptake process was ignored.

111 **3 Experimental results**

112 *3.1 Swelling pressure kinetics*

113 The evolution of swelling pressure for samples with different bentonite fractions and dry densities is
114 presented in Fig. 1. On the whole, for the samples with high bentonite fractions (larger than 50%)
115 and high dry densities (larger than 1.50 Mg/m^3), the swelling pressure increased rapidly and then
116 reached stabilization. For the samples with low bentonite fractions (smaller than 20%) and low dry
117 densities (smaller than 1.78 Mg/m^3), the swelling pressure started with a fast increase followed by a
118 peak value, a decrease and then reached stabilization. The presence of these peaks is related to the
119 collapse of macro-pores between soil grains (Pusch, 1982; Komine and Ogata, 1994). The lower the
120 dry density, the larger the volume of the macro-pores among grains (Lloret and Villar, 2007). At the
121 same dry density, more macro-pores are expected in the samples with larger fractions of claystone,
122 because of the higher unit mass of claystone (2.31 Mg/m^3) as compared with that of bentonite (2.00
123 Mg/m^3). It can also be observed that the time required to reach stabilization decreased with the

124 decrease of dry density and bentonite fraction, which can be explained by the higher hydraulic
125 conductivity of samples with a lower dry density and a larger claystone fraction.

126 *3.2 Relationship between final swelling pressure and dry density of the mixture*

127 Fig. 2 depicts the changes of final swelling pressure (P_s) with the dry density of samples. The results
128 of Karnland et al. (2008) on pure MX80 bentonite, Tang et al. (2011) on pure COx claystone, Wang
129 et al. (2012) and Zhang and Kröhn (2019) on MX80 bentonite/COx claystone mixture are also
130 presented. The swelling pressure values remarkably agree even though the tested materials were from
131 different batches, suggesting that the influence of MX80 bentonite and claystone batches was not
132 enough as to be relevant in the swelling pressure tests. For a given bentonite fraction, the final
133 swelling pressure increased with increasing dry density. At the same dry density, the final swelling
134 pressure of bentonite/claystone mixture after saturation was much lower than that of pure bentonite,
135 indicating that the addition of claystone reduced the global swelling capability.

136 **4 Interpretation and discussion**

137 *4.1 Contribution of claystone to swelling pressure*

138 As shown in Fig. 3a, each bentonite-based mixture after the full bentonite hydration process can be
139 divided into four parts: bentonite, voids in bentonite, additive (crushed COx claystone or sand) and
140 voids in additive (Wang et al., 2012; Deng et al., 2017). The bentonite dry density (ρ_{db}) can be
141 formulated by the following equation:

$$142 \quad \rho_{db} = \frac{(B/100)\rho_m G_{sa}\rho_w}{G_{sa}\rho_w(1+w_m/100)-\rho_m(1-B/100)(1+G_{sa}w_a)} = \frac{(B/100)\rho_{dm} G_{sa}\rho_w}{G_{sa}\rho_w-\rho_{dm}(1-B/100)(1+G_{sa}w_a)} \quad (1)$$

143 where ρ_m (Mg/m^3) is the mixture density; ρ_{dm} (Mg/m^3) is the dry density of the mixture; ρ_w is the
144 water unit mass; B (%) is the bentonite fraction (in dry mass) in the mixture; w_m is the water content
145 of the mixture; w_a is the water content of additive; G_{sa} is the specific gravity of additive.

146 For bentonite/sand mixtures, the water content of inactive sand is regarded as zero and the
147 bentonite dry density can be calculated directly using Eq. (1). A unique relationship has been
148 identified between swelling pressure and dry density of Avonseal (Gray et al., 1984), Calcigel (Agus
149 and Schanz, 2008), GMZ (Cui et al., 2012) and MX80 (Saba, 2013) bentonites regardless of the
150 bentonite fraction. For the MX80 bentonite/claystone mixture, the bentonite dry density of samples
151 cannot be calculated directly since the water content of claystone after saturation is unknown. In
152 terms of swelling pressure, the swelling capacity of pure bentonite is more than 100 times larger than
153 that of pure claystone at the same dry density (Fig. 3b) and the initial suction of pure bentonite is
154 about 4 times higher than that of pure claystone. During wetting, the bentonite in the mixture swelled
155 rapidly and came in full contact with the claystone while the volume change of the claystone was
156 influenced by the swelling pressure from bentonite. To determine the bentonite and claystone dry
157 densities, the final pressure at the interface of bentonite and claystone grains was assumed to be
158 equal to the global swelling pressure. This assumption was also adopted by Yang et al. (2002) when
159 investigating the consolidation behavior of lumpy granular soil under one-dimensional condition. In
160 this case, the bentonite dry density in the mixture can be estimated according to the relationship
161 between the swelling pressure of pure bentonite and its dry density (Fig. 3b):

$$162 \quad P_s = 1.652 \times 10^{-4} \exp^{6.781\rho_{db}} \quad (2)$$

163 At a certain global swelling pressure P_s , the corresponding bentonite dry density ρ_{db} in the
164 bentonite/claystone mixture can be back-calculated through the unique relationship for pure
165 bentonite. The claystone dry density ρ_{dc} in the mixture, the ratio of solid mass of claystone to the
166 volume occupied by claystone (Fig. 3a), can be calculated using Eq. (3):

167
$$\rho_{dc} = \frac{m_{sc}}{V - V_b} = \frac{V\rho_{dm}(1-B/100)}{V - \frac{V\rho_{dm}(B/100)}{\rho_{db}}} = \frac{\rho_{db}\rho_{dm}(1-B/100)}{\rho_{db} - \rho_{dm}(B/100)} \quad (3)$$

168 where m_{sc} is the solid mass of claystone; V is the total volume of the mixture; V_b is the volume of
169 bentonite.

170 Then, the claystone void ratio e_c in the mixture can be deduced using Eq. (4):

171
$$e_c = \frac{G_{sc}\rho_w}{\rho_{dc}} - 1 \quad (4)$$

172 where G_{sc} is the specific gravity of claystone. The calculated e_c is summarized in Table 3.

173 If the claystone can swell without the influence of bentonite, the expected claystone dry density
174 ρ_{dc}^e needed to achieve the above swelling pressure P_s can be similarly obtained from the correlation
175 between the swelling pressure of pure claystone and its dry density (Fig. 3b) :

176
$$P_s = 2.208 \times 10^{-8} \exp^{8.802\rho_{dc}^e} \quad (5)$$

177 Correspondingly, the expected claystone void ratio e_c^e can be deduced using Eq. (6):

178
$$e_c^e = \frac{G_{sc}\rho_w}{\rho_{dc}^e} - 1 \quad (6)$$

179 During wetting, the stress states of claystone grains in bentonite/claystone mixture and pure
180 claystone are different. In the former, the wetting of claystone is under the pressure imposed by the
181 swelling of bentonite and the water absorption of claystone is restrained (Attom and Barakat, 2000).
182 By contrast, in the latter, the pressure between claystone grains gradually increases with wetting.
183 Therefore, at a given global swelling pressure, e_c in the bentonite/claystone mixture should be
184 smaller than e_c^e , which is corroborated by the calculated result in Table 3. The difference between e_c
185 and e_c^e physically represents the inhibition degree of bentonite on the swelling of claystone. On the
186 whole, the larger the bentonite fraction, the larger the difference and the greater the inhibition effect.

187 For further analysis of the interaction between bentonite and claystone in swelling pressure

188 development, an inhibition factor η is defined as follows:

$$189 \quad \eta = \frac{e_c^e - e_c}{e_c^e} = 1 - \frac{e_c}{e_c^e} \quad (7)$$

190 The calculated inhibition factor values for the samples tested are summarized in Table 3. Fig. 4a
191 depicts the variation of η with bentonite fraction. For pure bentonite, the value of inhibition factor is
192 1; for pure claystone, it equals 0. The inhibition factor increases with the increasing bentonite
193 fraction, following a nonlinear relationship. This relationship can be well described by the following
194 expression with a squared correlation coefficient $R^2 = 0.972$:

$$195 \quad \eta = \frac{1}{1 + e^{10.664(0.595 - B/100)}} \quad (8)$$

196 From Fig. 4a, two inflection points of the sigmoidal curve can be identified, at approximately 40%
197 and 70% bentonite fractions. The inhibition effect of bentonite on the swelling of claystone can be
198 divided into three zones according to these two critical values, that is, Zone I, $100 \geq B \geq 70$; Zone II,
199 $70 > B \geq 40$; Zone III, $40 > B \geq 0$. Fig. 4b shows the sketch of bentonite/claystone mixture before and
200 after full saturation in the three zones.

201 In Zone I, $100 \geq B \geq 70$, the inhibition factor η is larger than 0.73. The fully swollen bentonite
202 grains form a matrix and claystone grains disperse into the matrix. The claystone grains behave as an
203 inert material and their swelling is almost completely inhibited by bentonite. After wetting, the
204 claystone void ratio e_c in the mixture (Table 3) is even slightly less than the initial void ratio of
205 claystone grain (0.17), due to the collapse of claystone under the swelling pressure induced by
206 bentonite.

207 In Zone II, $70 > B \geq 40$, the inhibition factor η is between 0.11 and 0.73. The swelling of
208 claystone grains is partially restrained by bentonite and the claystone can swell in volume and fill up
209 some voids in the mixture. In this case, partially swelling of the claystone will contribute to the

210 global swelling pressure.

211 In Zone III, $40 > B \geq 0$, the inhibition factor η is smaller than 0.11. Both claystone and bentonite
212 grains swell upon wetting. The fully swollen bentonite grains disperse in the skeleton formed by the
213 swollen claystone. The claystone void ratio e_c in the mixture is very close to the expected claystone
214 void ratio e_c^e , suggesting that the global swelling pressure is governed by claystone.

215 *4.2 Estimation of swelling pressure of bentonite/claystone mixture*

216 According to the inhibition factor determined above and the relationships between swelling pressure
217 and dry density for pure bentonite and claystone, the swelling pressure of samples with different
218 bentonite fractions and dry densities can be computed following an iteration procedure. Fig. 5 is a
219 flowchart showing how to predict the swelling pressure of MX80 bentonite/COx claystone mixture
220 in the full range of bentonite fraction. The general outline of this procedure is summarized as follows:

- 221 (a) to assign an initial value of swelling pressure P_0 ;
- 222 (b) to back-calculate the expected claystone dry density based on Eq. (5) and to determine the
223 corresponding expected claystone void ratio using Eq. (6);
- 224 (c) to determine the inhibition factor using Eq. (8) and to compute the claystone void ratio in the
225 bentonite/claystone mixture using Eq. (7);
- 226 (d) to apply Eq. (4) to calculate the claystone dry density and to back-calculate the bentonite dry
227 density using Eq. (3);
- 228 (e) to compute the swelling pressure P_1 according to Eq. (2);
- 229 (f) to judge the absolute error between P_1 and P_0 : if $|P_1 - P_0|$ is smaller than a certain tolerance α
230 (0.0001 MPa in this study), P_1 is the predicted value of swelling pressure; else, to assign $P_0 = P_1$ and
231 to go to step (a) for a new iteration.

232 The predicted and the measured swelling pressures are compared in Fig. 2, as a function of the
233 dry density of the mixture, showing a good agreement. This agreement shows the performance of the
234 proposed method as well as the relevance of the identified swelling mechanism.

235 **5 Conclusions**

236 The swelling pressure of MX80 bentonite/COx claystone mixture with different bentonite fractions
237 was investigated by carrying out constant-volume swelling pressure tests. The obtained results allow
238 the following conclusions to be drawn:

239 The swelling of claystone in the mixture can be inhibited by bentonite and the contribution of
240 claystone to the global swelling pressure depends on the bentonite fraction (B). An inhibition factor
241 was introduced as a function of the bentonite fraction to describe the inhibition effect. According to
242 two inflection points (40% and 70%), the inhibition effect can be divided into three zones (Zone I,
243 $100 \geq B > 70$; Zone II, $70 \geq B > 40$; Zone III, $40 \geq B \geq 0$). In Zone I, the swelling of claystone is almost
244 totally inhibited by bentonite; in Zone II, the swelling of claystone grains is partially restrained by
245 bentonite and the claystone can swell and contribute to the global swelling pressure of the mixture; in
246 Zone III, claystone fully swells upon wetting and the claystone governs the global swelling pressure.

247 A predictive method was proposed, allowing the swelling pressure of the mixture with different
248 bentonite fractions and dry densities to be calculated. The good agreement between the calculated
249 and measured swelling pressure values showed the performance of the proposed method as well as
250 the relevance of the identified swelling mechanism.

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Table 1 Physical and chemical properties of MX80 bentonite (data from Tang et al. (2008), Saba (2013) and Wang et al. (2014))

Soil property	Description
Specific gravity	2.76-2.77
Consistency limit	
Liquid limit (%)	520-575
Plastic limit (%)	42-53
Plasticity index (%)	478-522
Cation exchange capacity (CEC) (meq/100 g)	78-85
Main exchangeable cations (meq/100 g)	
Na ⁺	60-67
K ⁺	1
Mg ²⁺	3-4
Ca ²⁺	5-8
Main minerals	
Montmorillonite (%)	70-92
Quartz (%)	3-15

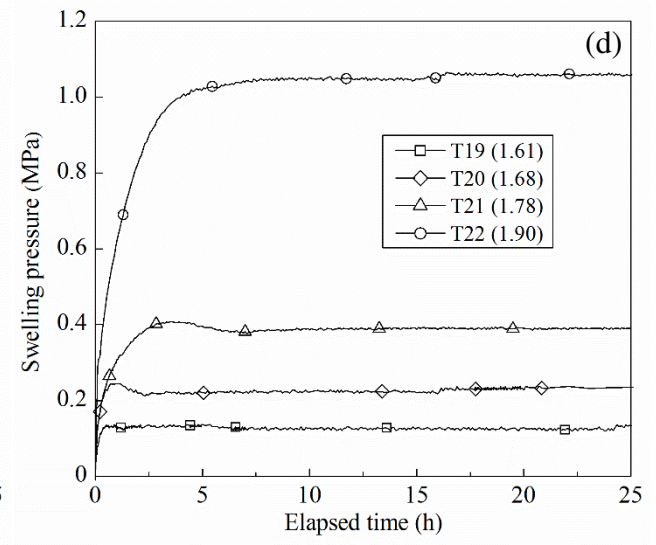
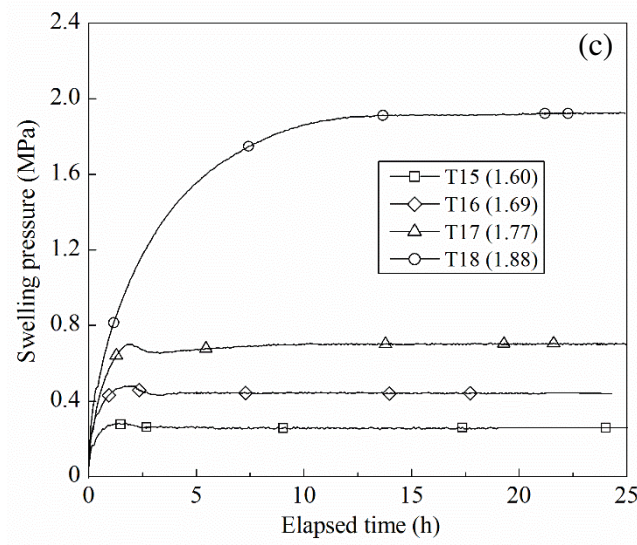
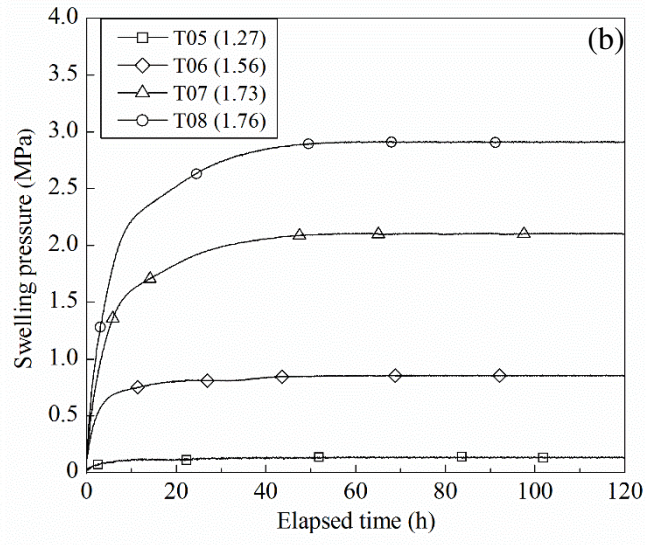
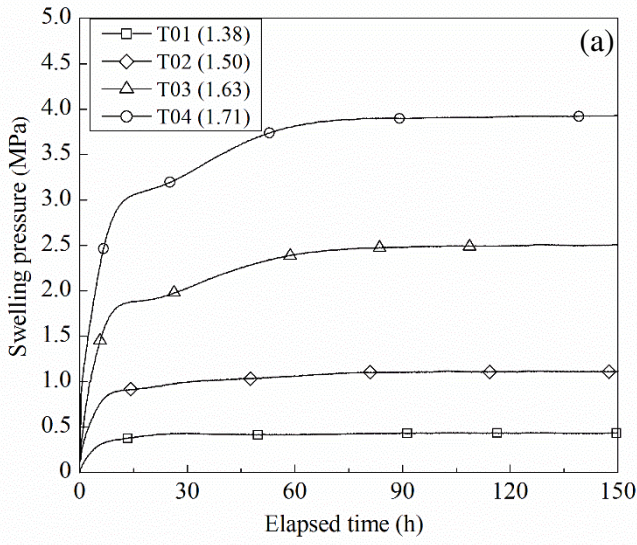
346 **Table 2** Receipt for preparing the synthetic water

Components	NaCl	NaHCO ₃	KCl	CaSO ₄ •2H ₂ O	MgSO ₄ •7H ₂ O	CaCl ₂ •2H ₂ O	Na ₂ SO ₄	Total
Content (g/L)	1.950	0.130	0.035	0.630	1.020	0.080	0.700	4.545

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Table 3 Test program and main results

Test No.	Bentonite fraction B (%)	Dry density of sample ρ_{dm} (Mg/m ³)	Initial water content w_m (%)	Bentonite dry density ρ_{db} (Mg/m ³)	Claystone void ratio e_c	Expected claystone void ratio e_c^e	Inhibition factor η	Final swelling pressure (MPa)	P_s
01	70	1.38	9.8	1.15	0.08	0.42	0.81	0.43	
02	70	1.50	9.8	1.29	0.13	0.34	0.60	1.11	
03	70	1.63	9.8	1.42	0.07	0.28	0.74	2.53	
04	70	1.71	9.8	1.48	0.02	0.25	0.91	3.94	
05	50	1.27	8.8	0.98	0.49	0.52	0.06	0.13	
06	50	1.56	8.8	1.26	0.30	0.36	0.16	0.85	
07	50	1.73	8.8	1.39	0.18	0.29	0.38	2.10	
08	50	1.76	8.8	1.44	0.19	0.27	0.28	2.91	
09	30	1.50	7.7	1.06	0.47	0.47	0.00	0.22	
10	30	1.60	7.7	1.16	0.41	0.41	0.00	0.46	
11	30	1.68	7.7	1.24	0.36	0.37	0.02	0.78	
12	30	1.79	7.7	1.35	0.30	0.31	0.06	1.59	
13	30	1.89	7.7	1.43	0.23	0.28	0.18	2.72	
14	30	1.99	7.7	1.52	0.18	0.23	0.23	5.23	
15	20	1.60	7.2	1.08	0.49	0.46	-0.06	0.26	
16	20	1.69	7.2	1.16	0.41	0.41	0.01	0.44	
17	20	1.77	7.2	1.23	0.35	0.38	0.06	0.70	
18	20	1.88	7.2	1.38	0.31	0.30	-0.02	1.93	
19	10	1.61	6.6	0.98	0.56	0.52	-0.08	0.14	
20	10	1.68	6.6	1.06	0.50	0.47	-0.07	0.23	
21	10	1.78	6.6	1.14	0.42	0.42	0.01	0.39	
22	10	1.90	6.6	1.29	0.35	0.34	-0.02	1.07	
23	0	1.80	6.1	-	-	-	0	0.15	
24	0	1.90	6.1	-	-	-	0	0.47	
25	0	1.99	6.1	-	-	-	0	0.79	

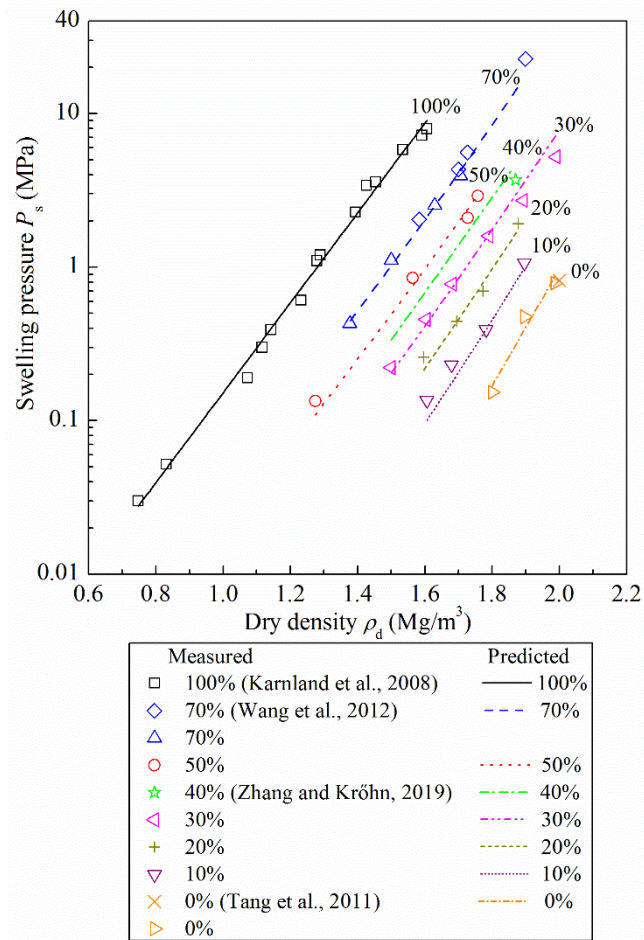


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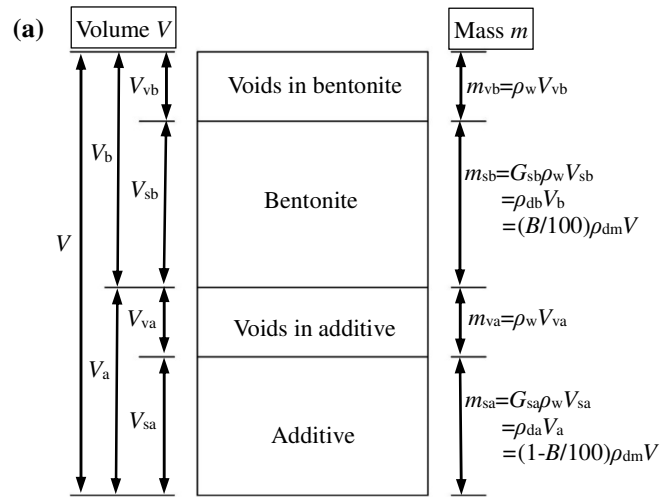
Fig. 1. Evolution of swelling pressure of bentonite/claystone mixture with bentonite fractions of (a) 70%; (b) 50%; (c) 20%; (d) 10%



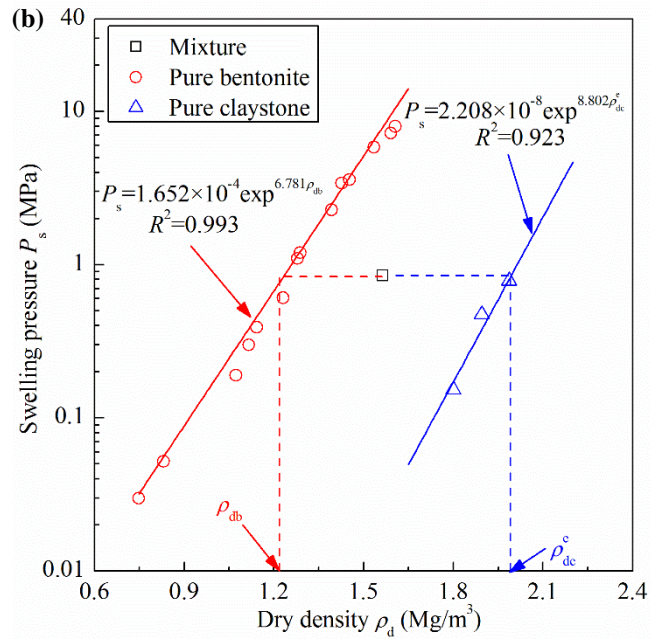
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Fig. 2. Measured and predicted swelling pressures as function of the dry density of samples. Note: the legend indicates the bentonite fraction

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Fig. 3. Composition of bentonite/additive mixture (a) and calculation of bentonite dry density and expected claystone dry density of bentonite/claystone mixture (b)

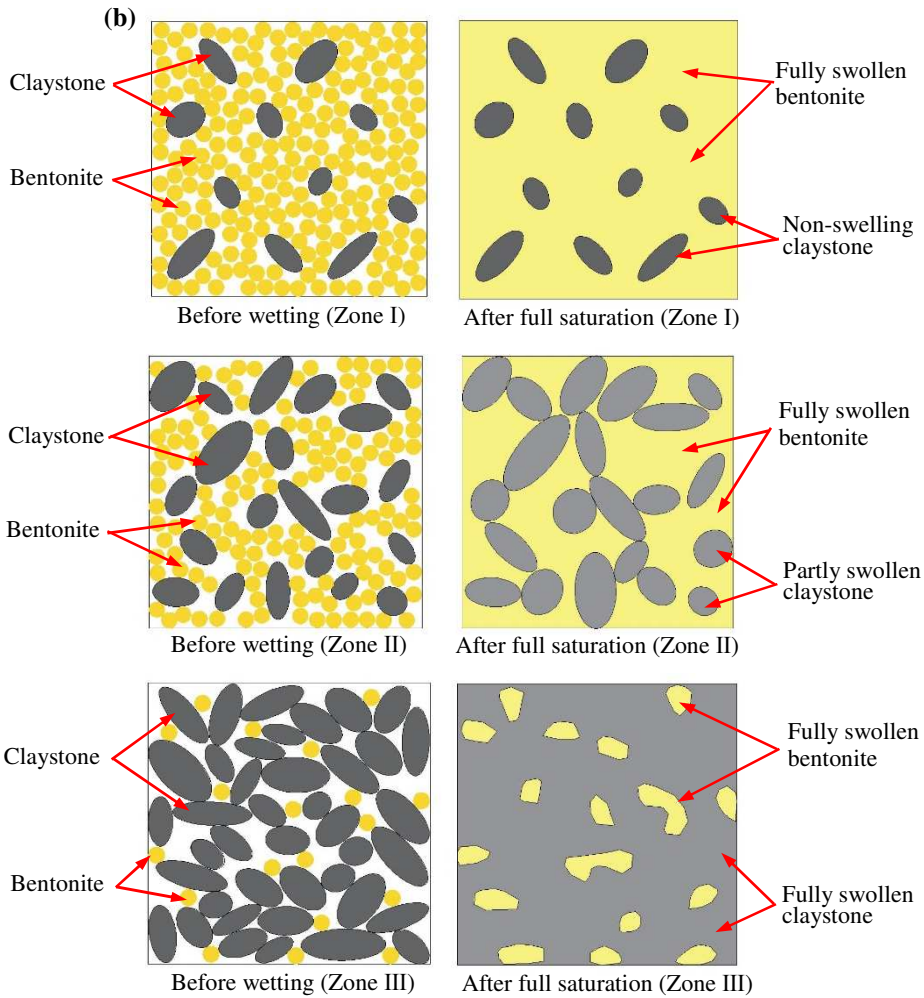
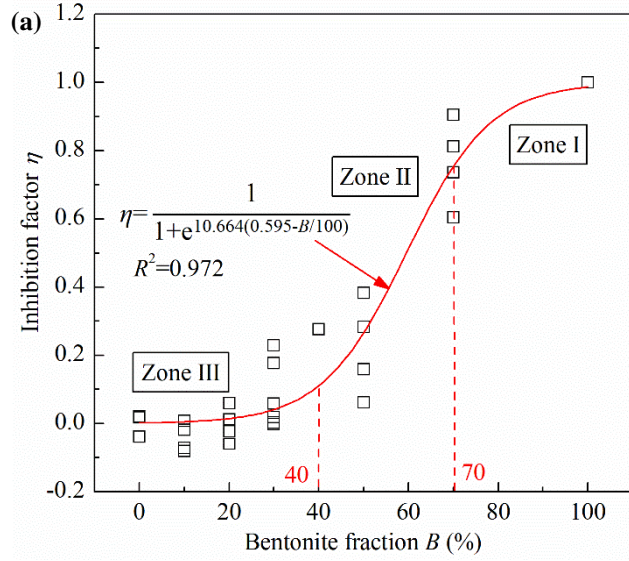


Fig. 4. Evolution of inhibition factor with bentonite fraction (a) and swelling mechanism of bentonite/claystone mixture in three zones (b). Note: the zones for bentonite and claystone after full saturation include voids in them

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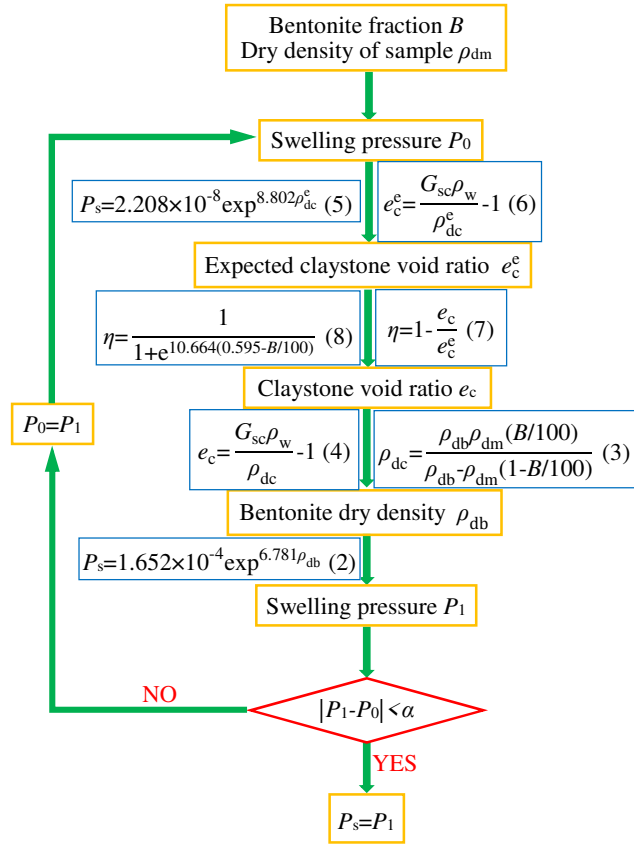


Fig. 5. Flowchart for swelling pressure prediction for bentonite/claystone mixture in the full range of bentonite fraction