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1 Linkage between swelling pressure, total suction of 2 saturated bentonites and suction of saturating aqueous 3 solutions

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10 **Abstract:** In deep geological repositories, compacted bentonites have been proposed
11 for use as barrier materials for isolating nuclear waste. The prevailing hydro-mechanical-
12 chemical boundary conditions in the repositories can affect the swelling capacity of compacted
13 bentonites. The present study examines the linkage between the swelling pressure of saturated
14 bentonites, the total suction of saturated bentonites and the suction of hydrating fluids. An
15 equation describing the linkage between these parameters was derived based on the thermo-
16 dynamics of soil moisture. To validate the derived relationship, laboratory tests involving
17 constant volume swelling pressure tests and total suction measurements after the swelling
18 pressure tests were carried out on a selected Ca-Mg-rich bentonite. Seven bentonite samples
19 with compaction dry density ranging from 1.20 and 1.83 Mg/m³ were hydrated with deionized
20 water, whereas four samples with a dry density of 1.59 Mg/m³ were hydrated with 0.0001 to
21 2.0 M CaCl₂ solutions. The total suctions of the saturated bentonite and the hydrating fluids
22 were determined by using a chilled-mirror hygrometer. The total suction of the saturated ben-
23 tonite was found to be affected by the compaction dry density and the suction of the hydrating
24 fluids. The experimental results from the present study and from the literature demonstrated
25 that the total suction of saturated bentonites minus the suction of the hydrating fluid used to
26 saturate bentonites is approximately equal to the swelling pressure of saturated bentonites.

27 **Keywords:** Expansive soils; bentonite; swelling pressure; suction; thermodynamics of soil
28 moisture; radioactive waste disposal

29 **1. Introduction**

30 A deep geological repository for the disposal of high-level radioactive wastes (HLWs)
31 may be located either in crystalline rock, argillaceous rock, or even in salt rock. The HLW
32 canisters isolated by compacted bentonites are planned to be placed in the disposal boreholes
33 located at a depth of 300 to 1000 m, depending on the disposal concepts (Pusch and Yong,
34 2006). Compacted bentonites are expected to swell upon imbibing groundwater thereby filling
35 the construction gaps and finally exerting a pressure on the host rock. Under this confined
36 condition and given the high installation dry densities, the swollen bentonites would possess
37 low porosity and low hydraulic conductivity. The saturated bentonite barriers are expected
38 to adsorb the positively charged radioisotopes from the HLWs and retard their migration to
39 the geosphere (Bergaya et al., 2006).

40 The swelling pressure of compacted bentonites has been considered as an important
41 mechanical parameter for assessing the long-term stability of the barrier in deep geological
42 repositories. From the perspective of laboratory measurements, the swelling pressure can
43 be defined as the pressure required to hold a bentonite sample at constant volume when it
44 absorbs fluid and tends to swell. Several investigations in the past have shown that the solute
45 concentration in the hydrating fluid, compaction dry density, and properties of bentonites
46 affect the magnitude of swelling pressure and the water content at saturation (Pusch, 1980*b*;
47 Bucher and Max, 1989; Pusch et al., 1990; Komine and Ogata, 1996; Karnland et al., 2005;
48 Castellanos et al., 2008; Villar and Lloret, 2008*b*; Schanz and Tripathy, 2009; Zhu et al., 2013;
49 Yigzaw et al., 2016).

50 The total suction of unsaturated soils consists of matric and osmotic suctions (Yong
51 and Warkentin, 1975; Fredlund et al., 2012; Lu and Likos, 2004). In clays, the matric suction

52 is due to surface tension, adsorptive forces and osmotic forces (i.e. the diffuse double layer
53 forces), whereas the osmotic suction is due to the presence of dissolved solutes in the pore
54 water.

55 Following saturation of a compacted bentonite sample with deionized water, the matric
56 and the total suction of the sample are generally assumed to be nearly equal to zero (e.g.
57 Wang et al. (2012), Agus et al. (2013), Yigzaw et al. (2016), Mäsin and Khalili (2016), among
58 others). Although the suction of deionized water is approximately equal to zero, however,
59 the assumptions with regard to a zero matric suction and a zero total suction in case of
60 saturated bentonites may not be considered appropriate. Fredlund et al. (2012) stated that
61 even when a soil is saturated under positive pore-water pressure, the osmotic component of
62 the total suction still remains. Tripathy et al. (2014a) found that the calculated osmotic
63 suction in various bentonites increased from 2 MPa to more than 19 MPa during the drying
64 process, depending upon the nature of exchangeable cations and the water content. Total
65 suction measurements after constant volume swelling pressure tests on a Na-bentonite and on
66 sand-bentonite mixtures that were saturated with deionized water and salt solutions have been
67 reported by Karnland et al. (2005) and Arifin (2008), respectively. These studies demonstrated
68 that the total suctions after the release of confinement in Na-bentonite and sand-bentonite
69 mixtures can be several hundreds of kPa to ten of MPa. The studies above imply that the
70 type and amount of exchangeable cations and the water content or the swollen dry density
71 influence the total suction of saturated bentonites.

72 In addition to the water content, temperature and composition of the pore water, the
73 overburden pressure influences the swelling clay suction (Croney, 1952; Kassiff and Shalom,
74 1971; Townr, 1981). They stated that for saturated swelling clays, the overburden pressure
75 was equal to the sum of effective suction and pore water pressure. The effective suction is
76 the matric suction of an unloaded sample of the soil whose water content is the same as
77 that of a loaded sample. The pore water pressure is the pore pressure of the loaded sample.

78 Moreover, Lambe and Whitman (1969); Nagaraj et al. (1994); Singhal et al. (2015) indicated
79 that removing overburden pressure from clays can induce a negative pore water pressure or
80 capillary suction.

81 A review of the literature presented herein suggested that an interdependency between
82 the swelling pressure, the total suction and the suction of hydrating fluids are expected in
83 case of swelling clays. Karnland et al. (2005) indicated that for saturated Na-bentonite, the
84 swelling pressure was the difference of the potential pressure (total suction) and the external
85 osmotic pressure (suction of hydrating fluids). The theoretical linkage between the swelling
86 pressure, the total suction and the suction of the hydrating fluids introduced by Karnland
87 et al. (2005) was based on constant volume condition. However, the experimental validation
88 of their linkage corresponds to unconfined condition. The total suction determined after re-
89 leasing the confinement conditions may be due to a negative pore water pressure or capillary
90 suction induced by unloading (Lambe and Whitman, 1969; Nagaraj et al., 1994; Singhal et al.,
91 2015). According to the effective stress concept, unloading would induce a negative pore wa-
92 ter pressure or capillary suction. Moreover, no systematic studies were made in the past to
93 explore the linkage between the swelling pressure, the total suction of saturated Ca-bentonite
94 and the suction of saturating aqueous solutions. Therefore, it is of interest to systematically
95 study the linkage between swelling pressure, total suction of saturated bentonites and suction
96 of saturating aqueous solutions. Understanding of the linkage between the various relevant
97 parameters will enable researchers to develop models that will facilitate evaluating the influ-
98 ence of hydro-mechanical-chemical boundary conditions on the stability of the barrier in deep
99 geological repositories.

100 The objective of this study was to examine the linkage between the swelling pressure,
101 the total suction and the suction of hydrating fluids in case of saturated bentonites. An
102 equation describing the linkage between these three parameters was first derived based on
103 the thermodynamics of soil moisture. Further, constant volume swelling pressure tests and

104 total suction measurements after the swelling pressure tests were carried out on a selected
105 bentonite. Based on the experimental results from this study and from the literature, the
106 derived equation was then validated.

107 **2. Theoretical considerations**

108 Total potential of soil-water has been discussed from the point of view of thermody-
109 namic considerations of soil moisture by numerous researchers (Edlefsen and Anderson, 1943;
110 Low and Deming, 1953; Bolt and Miller, 1958; Mitchell, 1962; Yong and Warkentin, 1975;
111 Iwata et al., 1995). Iwata et al. (1995) stated that the total water potential (μ) in clays rela-
112 tive to the potential of the free pure water at the same temperature and external pressure is
113 comprised of the potentials due to surface tension (μ_c), van der Waals force (μ_f), solutes (μ_0),
114 electric field (μ_e), temperature (μ_T), pressures (μ_P), and gravitational field (μ_g) (Eq. 1).

$$\mu = \mu_c + \mu_f + \mu_0 + \mu_e + \mu_T + \mu_P + \mu_g \quad (1)$$

115 Detailed description about these potentials could be found in the work of Iwata et al. (1995).
116 Since μ_c , μ_f , μ_0 , μ_e and μ_P are the potentials which are closely related the discussion in this
117 paper, their definitions are given in the following paragraphs.

118 Curved surfaces of water are formed in soils pores under unsaturated condition. The
119 water menisci give rise to a decrease in the total potential of the water and this decrease is
120 described by the surface tension effect or capillary potential (μ_c).

121 Van der Waals force effect (μ_f) corresponds to the potential energy of interaction be-
122 tween clay surface and a water molecule. Therefore, μ_f results in a decrease in the total
123 potential of the water and the decrease in the total potential of soil water depends upon the
124 distance from the surface of the clay particle to a water molecule.

125 Solute effect (μ_0) results from various solutes in the soil solution. The various solutes
126 in the soil solution result in a decrease in the total potential of soil water and the decrease in

127 the total potential of soil water depends on the concentration of the various solutes in the soil
128 solution.

129 Clay minerals have electric charge and the soil solution is affected by the electric field
130 induced by those charges. The change of the total potential of water in soil because of electric
131 displacement is defined as μ_e .

132 To satisfy the requirement of equilibrium, the concept of the effect of pressures (μ_P)
133 is introduced. According to the thermodynamics of soil moisture, at equilibrium state, the
134 effect of pressures compensates for the drop in the total potential of the water near the clay
135 particle caused by surface tension, van der Waals force, solutes, electric field, temperature and
136 gravitational field.

137 The applicability of the potentials above can be examined in the light of a constant
138 volume swelling pressure test on compacted bentonite. Studies on the pore size distribution of
139 compacted bentonite-based materials have shown dual-structure pore size distributions with
140 inter-aggregate pores and intra-aggregate pores (Sridharan et al., 1971; Lloret et al., 2003;
141 Delage et al., 2006; Romero and Simms, 2008; Seiphoori et al., 2014). The inter-aggregate
142 pores and intra-aggregate pores are generally identified with macrostructural porosity and
143 microstructural porosity, respectively, when analysing compacted clays (Alonso et al., 1990;
144 Gens and Alonso, 1992; Musso et al., 2013; Manca et al., 2015; Navarro et al., 2017a, 2018).
145 Similarly, the total potential of macrostructural pore water (μ_M) and the total potential of
146 microstructural pore water (μ_m) have been considered in this work to analyse the constant
147 volume swelling pressure tests on compacted bentonite.

148 Sketches of a swelling pressure test device and of the structure of compacted bentonite
149 (macro and micro structure) are shown in Fig. 1. A compacted bentonite sample under
150 constant volume condition is hydrated with an aqueous solution. The upon saturation of the
151 sample in the measuring system (Fig. 1) during a swelling pressure test, the macrostructural
152 pores are filled with the aqueous solution. In addition, the microstructural pores are also filled

153 with water that may also include some solutes. At saturation, the water potential caused by
 154 surface tension (μ_c) gets eliminated. Considering that the temperature and the atmospheric
 155 pressure change little during an isothermal swelling pressure test, the potentials caused by
 156 temperature (μ_T) and atmospheric pressure can be neglected. Since the scale of this system
 157 is very small, it is reasonable to neglect the gravitational potential (μ_g). Therefore, the total
 158 potential of the macrostructural pore water (μ_M) would be reduced to only the potential due
 159 to solutes (μ_0^M) and is given by Eq. (2). In a swelling pressure test, μ_0^M corresponds to the
 160 total water potential of the hydrating fluid (μ_a). In suction terms, the total suction of the
 161 hydrating fluid is ψ_a . In Eq. (2), \bar{v}_w is the partial specific volume of water.

$$\mu_M = \mu_a = -\psi_a \bar{v}_w \quad (2)$$

162 Similarly, the total potential of the microstructural pore water (μ_m) is given by Eq. (3). P
 163 is the pressure compensating for the drop in the total potential of the water near the clay
 164 particle due to van der Waals force, solutes and electric field and is approximately equal to
 165 net mean stress.

$$\mu_m = \mu_f^m + \mu_0^m + \mu_e^m + P\bar{v}_w \quad (3)$$

166 According to the thermodynamics of soil moisture, at equilibrium state, the total po-
 167 tential of the water at any given points in the system has a constant value. Thus, Eq. (4) can
 168 be obtained.

$$\mu_m = \mu_M \quad (4)$$

169 By rearranging Eqs. (2), (3), and (4), Eq. (5) can be obtained:

$$\mu_f^m + \mu_e^m + \mu_0^m = -\psi_a \bar{v}_w - P\bar{v}_w \quad (5)$$

170 It is generally assumed that \bar{v}_w does not depend on pressure and is a constant, nearly $1 \text{ m}^3/\text{Mg}$.

171 A simplification of Eq. (5) yields Eq. (6).

$$-\frac{1}{\bar{v}_w}(\mu_f^m + \mu_e^m + \mu_0^m) - \psi_a = P \quad (6)$$

172 Since $(\mu_f^m + \mu_e^m)$ and μ_0^m correspond to the hydration effect and osmotic effect, respec-
 173 tively, thus they correspond to matric potential and osmotic potential, respectively. Because
 174 $(\mu_f^m + \mu_e^m + \mu_0^m)$ corresponds to the sum of matric and osmotic potentials in case of saturated
 175 clays, their value can be approximately determined from Kelvin's equation (i.e. Eq. (7)).

$$\mu_f^m + \mu_e^m + \mu_0^m = \frac{RT}{M_w} \ln(RH) \quad (7)$$

176 In Eq. (7), R is the molar gas constant, M_w is the molecular mass of water, and RH is the
 177 relative humidity. In suction terms, the total suction (ψ_t) of the saturated bentonite under
 178 constant volume condition is given in Eq. (8)

$$\psi_t = -\frac{RT}{M_w \bar{v}_w} \ln(RH) \quad (8)$$

179 After Eqs. (6), (7), and (8) are combined, Eq. (9) can be obtained in suction and
 180 pressure terms, where P_s represents swelling pressure.

$$\psi_t - \psi_a = P = P_s \quad (9)$$

181 Croney (1952); Kassiff and Shalom (1971); Townenr (1981) suggested a similar relationship to
 182 Eq. (9) for a saturated clay under compression and no water content changes, i.e.

$$s + U = P \quad (10)$$

183 where s is the matric suction measured under zero load, corresponding to ψ_t in Eq. (9); U is
 184 the pore water pressure measured under load, corresponding to $-\psi_a$ in Eq. (9); P is the total
 185 external all-round pressure, corresponding to P_s in Eq. (9). If the aqueous solution in Fig. 1
 186 is pure water ($\psi_a = 0$), then Eq. (9) is reduced to the thermodynamic equations developed by
 187 Low and his co-workers (Low and Anderson, 1958; Viani et al., 1983; Low, 1987). In addition,
 188 similar expressions have been also given by Karnland et al. (2005) and Navarro et al. (2017a).

189 To validate Eq. (9) experimentally, separate measurements of P_s , ψ_a and ψ_t are neces-
 190 sary. The former two can be directly determined by a swelling pressure device and a chilled-
 191 mirror hygrometer, respectively. However, the latter cannot be directly determined by placing

192 a relative humidity sensor in a constant volume swelling pressure device. In constant volume
 193 condition, the water potential determined by the relative humidity sensor is the total water
 194 potential (μ_m or μ_M). This can be confirmed by constant volume infiltration experimental
 195 results reported by Cui et al. (2008); Ye et al. (2009); Mokni and Barnichon (2016); Villar
 196 et al. (2016). They found that the relative humidity above the wetting face reached nearly
 197 100% after long-term of supplying water. This indicates that the water potential determined
 198 by the relative humidity sensor in constant volume condition is equal to the water potential
 199 of the hydrating fluid (water). Thus, releasing swelling pressure by removing the confinement
 200 conditions is necessary for determining ψ_t or $(\mu_f^m + \mu_e^m + \mu_0^m)$.

201 If the hydrating fluid (Fig. 1) is removed before releasing the swelling pressure, subse-
 202 quent removal of swelling pressure can be considered as undrained unloading. Consequently,
 203 the water content of the saturated bentonite sample would not change after releasing the
 204 swelling pressure. Since μ_f^m and μ_e^m are dominated by water content or volume, it is reason-
 205 able to assume that they would not change after releasing the swelling pressure. Because μ_0^m
 206 is dominated by the molarity of solutes and the unloading does not change the mass of solutes
 207 and the water content, μ_0^m would remain after releasing the swelling pressure. This would be
 208 also true for μ_0^M . Thus, $(\mu_f^m + \mu_e^m + \mu_0^m)$ or ψ_t remains unchanged before and after releasing
 209 the swelling pressure.

210 Moreover, undrained unloading would induce negative pore water pressure or capillary
 211 suction ($u_a - u_w$) (Lambe and Whitman, 1969; Nagaraj et al., 1994; Singhal et al., 2015).
 212 Since the effective stress of a soil sample remains during undrained unloading, a decrease in
 213 the net mean stress would be balanced by an equal increase in $(u_a - u_w)$ (Lambe and Whitman,
 214 1969; Nagaraj et al., 1994). Thus, the total potential of the macrostructural pore water after
 215 releasing swelling pressure (μ_M^{un}) would change to the expression shown in Eq. (11).

$$\mu_M^{un} = \mu_0^M - (u_a - u_w)\bar{v}_w = \mu_0^M - P_s\bar{v}_w = -(\psi_a + P_s)\bar{v}_w \quad (11)$$

216 Because of the removal of the swelling pressure, the total potential of the microstruc-
217 tural pore water (μ_m^{un}) would be reduced to Eq. (12).

$$\mu_m^{\text{un}} = \mu_f^m + \mu_e^m + \mu_0^m = -\psi_t \bar{v}_w \quad (12)$$

218 Comparison of Eq. (11) and Eq. (12) indicates that the total potential of the macrostruc-
219 tural pore water would be still equal to the total potential of the microstructural pore water
220 after releasing the swelling pressure. In other words, the thermodynamic equilibrium remains
221 during undrained unloading, which would mean that there is no water exchange between the
222 macrostructural pore and the microstructural pore water or that the assumption of undrained
223 unloading is reasonable. Thus, after releasing the swelling pressure, the total suction of the
224 saturated bentonite would be equal not only to the capillary suction triggered by swelling pres-
225 sure release but also to the total suction (ψ_t) of the saturated bentonite under constant volume
226 condition. Consequently, ($\mu_f^m + \mu_e^m + \mu_0^m$) or ψ_t can be also determined by a chilled-mirror
227 hygrometer after releasing the swelling pressure.

228 To validate Eq. (9) experimentally, constant volume swelling pressure tests were per-
229 formed on a Ca-Mg-rich bentonite using various aqueous solutions, i.e. deionized water and
230 solutions of CaCl_2 . After the swelling pressure tests, the suctions (ψ_a) of the saturating aque-
231 ous solutions and the total suctions (ψ_t) of the saturated bentonite samples were determined.

232 3. Materials and methods

233 3.1. Materials

234 The bentonite used in this study was Calcigel bentonite from Bavaria, Germany and
235 purchased from Süd-Chemie AG Moosburg, Germany. The bentonite was composed of mont-
236 morillonite (60-70%), quartz (6-9%), feldspar (1-4%), kaolinite (1-2%), mica (1-6%), calcite
237 (2-4%), dolomite (1-3%) and other minerals (3%). The total specific surface area of the ben-
238 tonite was measured by the ethylene glycol monoethyl ether (EGME) method (Cerato and

239 Lutenegger, 2002) and was found to be $436 \text{ m}^2/\text{g}$. The total cation exchange capacity (CEC)
240 of the bentonite was $74 \text{ cmol}(+)/\text{kg}$. More than 85% of the exchangeable cations in the ben-
241 tonite were divalent calcium and magnesium ions and about 12% of the exchangeable cations
242 were monovalent sodium ions (Schanz and Tripathy, 2009; Baille et al., 2010). Based on the
243 measurements of osmotic suction using the squeezing technique (Fredlund et al., 2012), Arifin
244 and Schanz (2009) noted that the osmotic suction of Calcigel bentonite remained at about 0.05
245 MPa for a large range of water content. This indicated that Calcigel contains some soluble
246 ions that contribute to about 0.05 MPa osmotic suction. The liquid limit, plastic limit and
247 particle specific gravity of the bentonite were 119%, 45% and 2.80, respectively.

248 **3.2. Experimental methods**

249 **3.2.1. Sample preparation**

250 Bentonite-water mixtures were prepared at target water contents of 9% and 20% by
251 adding the required amounts of deionized water to the bentonite. After mixing bentonite and
252 water, the bentonite-water mixtures were kept in a two-layer plastic bag for about two weeks
253 for moisture equilibration. The water contents of the mixtures were determined by oven
254 drying method at a temperature of 105°C prior to preparing samples for swelling pressure
255 tests. The total suctions of the mixtures were determined by using AquaLab-3TE chilled-
256 mirror hygrometer (Decagon Devices, 1999-2009; Leong et al., 2003) and Kelvin's law (i.e.
257 Eq. (8)). The accuracy of the device in terms of the relative humidity was $\pm 0.3\%$. If the
258 total suction of a soil sample was equal to 0.3 MPa, the determined total suction could vary
259 from 0 to 0.8 MPa at 22°C due to the accuracy of $\pm 0.3\%$. If the total suction of a soil sample
260 was equal to 3.0 MPa, the determined total suction could vary from 2.6 to 3.4 MPa at 22°C .
261 This indicates that the accuracy of $\pm 0.3\%$ could cause a significant total suction measurement
262 error (i.e. up to $\pm 140\%$) if the total suction of a soil sample was less than 0.3 MPa.

263 Compacted bentonite samples, 50 mm in diameter and 15 ± 0.02 mm in height, were

264 prepared by statically compacting bentonite-water mixtures inside the oedometer ring by using
265 a 15-tonne capacity compression testing machine. The samples were compacted by applying
266 uniaxial loads. The initial compaction conditions of the as-compacted samples and aqueous
267 solutions used to saturate the samples during the swelling pressure tests are shown in Table
268 1.

269 **3.2.2. Determination of swelling pressure, water content and total suction**

270 An isochoric swelling pressure device (Fig. 1) (Romero, 1999; Schanz and Tripathy,
271 2009) was used in this study for measuring the swelling pressures of compacted bentonite
272 samples. After the static compaction process, the as-compacted sample inside the sample
273 ring was placed in the swelling pressure device. Samples CB-0 to CB-6 with compaction dry
274 densities varying between 1.20 to 1.83 Mg/m³ (Table 1) were hydrated by supplying deionized
275 water. Samples CB-2, CB-3 and CB-4 had very similar compaction conditions (dry density
276 = 1.57 Mg/m³ and water content = 20%). Samples CB-7, CB-8, CB-9, and CB-10 (dry
277 density = 1.59 Mg/m³) were hydrated with deionized water, 0.5, 1.0 and 2.0M CaCl₂ solutions,
278 respectively. The hydrating fluids were supplied from burettes attached to the bottom fluid
279 reservoirs of the devices. During the swelling pressure tests, the CaCl₂ solutions were renewed
280 periodically for maintaining the constant concentration of the solutions.

281 The swelling pressures tests for all samples were terminated once the swelling pressure
282 was found to be constant. Samples CB-0 and CB-4 were dismantled after about 17 and
283 19 days, respectively. For the other samples the testing duration well exceeded 30 days.
284 Following termination of a test, the fluid supply to the sample was stopped. Subsequently,
285 samples of the aqueous solution (i.e. the hydrating fluid) were collected (in duplicate) from
286 the bottom fluid reservoir for determining the suctions of the fluid. Before dismantling the
287 device, pressurized humid air (relative humidity = 100%) generated by deionized water was
288 used to flush the bottom and top fluid reservoirs to minimize fluid uptake by samples upon

289 unloading. Afterwards, the sample was removed from the isochoric device for determining
290 total suction (ψ_t) and water content.

291 In order to explore the homogeneity of samples in terms of the water content and
292 suction, the bentonite samples were cut into three parts that represent the top, middle and
293 bottom layers, respectively. The water contents and total suctions of the representative sam-
294 ples were determined by the oven drying method (105°C) and the chilled-mirror hygrometer,
295 respectively. Sufficient care was exercised to minimize the evaporation water from the samples
296 during preparing samples for water content and suction measurements.

297 **3.2.3. Determination of suctions of hydrating fluids/aqueous solutions**

298 The suction of the deionized water (kPa) used in this study was determined from the
299 electrical conductivity (EC) measurements (mS/cm) using Eq. (13) (Rao and Shivananda,
300 2005).

$$\psi_a = 38.54EC^{1.0489}. \quad (13)$$

301 The electrical conductivity (EC) of the deionized water used in this study was found to be
302 about 0.03 mS/cm. Thus, the calculated suction of the deionized water used was approximately
303 0.001 MPa and was nearly equivalent to the suction of a 0.0001 M CaCl₂ solution based on
304 van't Hoff equation. The suctions of CaCl₂ solutions after the swelling pressure tests were
305 determined by using the chilled-mirror hygrometer and Kelvin's law. The suctions of 0.5 M, 1
306 M and 2 M CaCl₂ solutions were found to be 3.03, 8.42, and 20.55 MPa, respectively. These
307 values were found to be similar to the suctions of the solutions prior to the swelling pressure
308 tests and remained within the measurement accuracy of the chilled-mirror hygrometer.

309 4. Results and discussion

310 4.1. Swelling pressure

311 The elapsed time versus swelling pressure for bentonite samples CB-0 to CB-7 is shown
312 in Fig. 2a. Similarly, Fig. 2b presents the evolution of swelling pressures in case of samples
313 CB-7, CB-8, CB-9, and CB-10 that were hydrated with deionized water, 0.5, 1.0 and 2.0
314 M CaCl₂ solutions, respectively. Swelling pressure equilibrium time was defined as the time
315 when the variation of swelling pressure started to be less than 5 kPa per day. The swelling
316 pressure equilibrium time was found to increase with an increase in the sample dry density
317 (Fig. 2a). The equilibrium time for sample CB-0 (dry density = 1.2 Mg/m³) was about 60
318 h, whereas it was found to be about 440 h for sample CB-6 with a compaction dry density of
319 1.83 Mg/m³. For a given dry density of 1.59 Mg/m³, the type of hydrating fluid (i.e., water
320 or CaCl₂ solutions) had no significant influence on the equilibrium time. In this case, the
321 swelling pressure equilibrated in about 270 h.

322 An increase in the swelling pressure equilibrium time with an increase in the dry density
323 is attributed to an increase in the static compaction pressure which in turn affects the pore-
324 size distribution and permeability of compacted bentonites (Pusch, 1980*a*; Lloret et al., 2003).
325 Studies on the pore size distribution of the compacted clays have shown dual-structure pore
326 size distributions with intra-aggregate and inter-aggregate pores (Sridharan et al., 1971; Pusch
327 and Yong, 2006; Delage et al., 2006). As the dry density increases, the inter-aggregate pore
328 size decreases, whereas the intra-aggregate pore size may remain almost constant (Lloret
329 et al., 2003; Romero and Simms, 2008; Li and Zhang, 2009; Wang et al., 2013; Seiphoori
330 et al., 2014). The flow of liquid water in compacted bentonites initially occurs within the
331 inter-aggregate pores and then in the intra-aggregate pores. Since the permeability of the
332 compacted bentonites is mainly governed by the inter-aggregate pores, a reduction in the size
333 of the inter-aggregate pores either due to an increase in compaction dry density or due to

334 swelling decelerates the development of swelling pressure.

335 The effect of sample dry density on the equilibrium swelling pressure is presented in
336 Figs. 3. The magnitude of swelling pressure at equilibrium was found to be influenced by the
337 sample dry density. As can be seen from Fig 3, with deionized water as the hydrating fluid
338 the swelling pressure increased from 0.3 to 10.5 MPa with an increase in the dry density from
339 1.20 to 1.83 Mg/m³. The differences in the swelling pressures of samples CB-2, CB-3, and
340 CB-4 (dry density = 1.57 Mg/m³) remained less than about 0.5 MPa. Based on the diffuse
341 double layer theory as applicable to interacting clay platelet systems, a dry density of 1.55
342 Mg/m³ for Calcigel bentonite delimits the osmotic swelling (Schanz and Tripathy, 2009). If
343 the dry density is less than the threshold dry density, diffuse double layer repulsion governs the
344 swelling pressure. An increase in the dry density decreases the distance between clay platelets,
345 increases the ion concentration difference between the central plane between clay platelets
346 and bulk solution and further increases the swelling pressure (Bolt, 1956; Van Olphen, 1977;
347 Schanz and Tripathy, 2009). If dry density is greater than the threshold dry density, hydration
348 forces control the swelling pressure. An increase in dry density decreases the space between
349 interlayers, increases hydration forces and further increases the swelling pressure (Van Olphen,
350 1977; Schanz and Tripathy, 2009).

351 In the case of samples with a constant dry density of 1.59 Mg/m³, the equilibrium
352 swelling pressure decreased with an increase in CaCl₂ concentration (Fig. 2b). A reduction in
353 the swelling pressure from 2.98 to 2.10 MPa was found to occur (i.e. a decrease of about 30%)
354 when the concentration of CaCl₂ was increased from 0.0001 to 2.0 M. Similar experimental
355 results concerning the influence of salt solution concentration on the swelling pressure have
356 been reported by several researchers (Pusch, 1980*b*; Bucher and Max, 1989; Pusch et al., 1990;
357 Karnland et al., 2005; Villar and Lloret, 2008*a*; Castellanos et al., 2008; Zhu et al., 2013;
358 Tripathy et al., 2014*b*).

359 4.2. Water content and total suction profiles

360 Figure 4 presents the profiles of the water content and the total suction for samples
361 CB-0 to CB-6 that were saturated with deionized water. Figure 5 presents the profiles of
362 the water content and the total suction for samples CB-7 to CB-10 that were saturated with
363 CaCl_2 solutions. Due to the water uptake by the samples during the swelling pressure tests,
364 the water contents of the samples at the end of the swelling pressure tests were found to be
365 higher than the initial water contents (Figs. 4a and 5a). For any sample, the final water
366 content was nearly the same along the depth of the sample. The degree of saturation of all
367 the samples tested was determined based on the final water contents, the dimensions of the
368 samples and the volume-mass relationships. The degree of saturation of all samples indicated
369 that all samples were fully saturated at the end of the swelling pressure tests.

370 As compared to the initial total suctions, the total suctions of all samples at the end of
371 the swelling pressure tests were found to have decreased (Figs. 4b and 5b). The total suction
372 profiles in most cases were found to be uniform. A variation of the total suction along the
373 depth of sample CB-4 was noted, in which case the total suction towards the top of the sample
374 was found to be greater than that occurred towards the bottom (Fig. 4b). The variation of the
375 total suction along the depth of sample CB-4 is attributed to a shorter testing time (19 days)
376 than for samples CB-2 and CB-3 that had similar compaction conditions, but were tested for
377 longer durations (more than 32 days).

378 The effect of dry density on the final water content and total suction of the samples
379 tested with deionized water is shown in Fig. 6a. The average values of the final water contents
380 and total suctions are plotted in Fig. 6a. With an increase in the dry density, the final water
381 content decreased and the total suction was found to increase.

382 The amount of water present in the saturated bentonite affects the total suction and
383 thus the magnitude of the total suction depends on the dry density of the saturated bentonite.

384 At high dry densities, the capillary effect disappears and the diffuse double layer is suppressed,
385 but the short-range adsorption effects (surface and ion hydration within the interlayers) remain
386 (Yong, 1999; Schanz and Tripathy, 2009). In this case, the matric suction caused by the short-
387 range adsorption effects contributes to the total suction and its magnitude increases with a
388 decrease in the thickness of the water layer within the interlayers. At low dry densities, the
389 matric suction due to the short-range adsorption effects diminishes, whereas the one due to the
390 diffuse double layer forces remains. In this case, the matric suction caused by diffuse double
391 layer forces contributes to the total suction and its magnitude increases with a decrease in the
392 thickness of the water layer within clay platelets (Yong, 1999; Tripathy et al., 2014a).

393 The concentration of CaCl_2 in the hydrating fluids affected the total suctions of samples
394 CB-7 to CB-10 (Fig. 6b). Since the samples had the same dry density, the saturated water
395 contents remained similar regardless of the concentration of the hydrating fluids. However,
396 the total suction increased significantly with an increase in the CaCl_2 concentration. With
397 an increase in the CaCl_2 concentration from 0.0001 to 2.0 M, the total suction increased from
398 3.2 to 22.7 MPa. The increase in the total suction with the CaCl_2 concentration is attributed
399 primarily to an increase in osmotic suction caused by an increase in the amount of dissolved
400 salts (CaCl_2) in the macrostructural pore fluid (Rao and Thyagaraj, 2007; Musso et al., 2013).

401 **4.3. Total suction and swelling pressure**

402 The experimental results presented in Figs. 3 and 6 show that both swelling pressure
403 and total soil suction were dependent on the dry density of the bentonite and salt concentration
404 of the hydrating fluids. Total suction versus swelling pressure plot is shown in Fig. 7. For
405 the bentonite samples that were hydrated with deionized water (samples CB-0 to CB-7), with
406 the exception of sample CB-0, the total suction was nearly equal to the swelling pressure. For
407 sample CB-0, the total suction (0.8 MPa) was greater than the swelling pressure (0.3 MPa),
408 which was probably the result of a significant total suction measurement error. As mentioned

409 in the section of Materials and methods, the total suction measurement error was up to $\pm 140\%$
410 if the total suction of a soil was less than 0.3 MPa. This significant error (i.e. $\pm 140\%$) could
411 have overestimated the real total suction of sample CB-0. In the case of the samples that were
412 hydrated with CaCl_2 solutions (samples CB-8 to CB-10), the total suction was considerably
413 greater than the corresponding swelling pressure. The test results showed that if the hydrating
414 fluid was water ($\psi_a = 0.001$ MPa), there was a good agreement between swelling pressure and
415 total suction. Disagreement between total suction and swelling pressure was only observed for
416 the samples that were hydrated with solutions having high salt concentration.

417 **4.4. Suction of hydrating fluids and total suction**

418 Figure 8 presents the suctions of the aqueous solutions used for hydrating the bentonite
419 samples versus the total suctions of all samples. As can be expected, the total suctions of the
420 samples that were saturated with deionized water (samples CB-0 to CB-7) far exceeded the
421 suction of water ($\psi_a = 0.001$ MPa). The total suction of the bentonite saturated with deionized
422 water increased from 0.8 MPa to 12.8 MPa with an increase in the dry density from 1.2 to 1.83
423 Mg/m^3 (Fig. 6a). For samples CB-8 to CB-10 that were saturated with CaCl_2 solutions, the
424 total suctions were found to be greater than the suctions of aqueous solutions, too. For the
425 bentonite saturated with CaCl_2 solutions, the total suction increased from 5.4 to 22.7 MPa
426 with an increase in the suction of CaCl_2 solutions from 3.0 to 20.6 MPa.

427 **4.5. Swelling pressure and difference between total suction and suc-** 428 **tion of hydrating fluids**

429 The swelling pressures (P_s) of saturated bentonites are plotted against the differences
430 between the total suctions of saturated bentonites and the suctions of the hydrating fluids
431 ($\psi_t - \psi_a$) in Fig. 9. In addition to the test results obtained from this study on the Ca-
432 Mg-rich bentonite, the test results reported by Karnland et al. (2005) on a Na-bentonite are

433 also shown in Fig. 9 for the tests with water and various NaCl solutions. In their study,
434 the compacted Na bentonite samples under constant volume condition were saturated with
435 solutions of concentration from 0.0001 to 3 M NaCl. After the swelling pressures reached
436 equilibrium, the samples were extracted from the constant volume device for total suction
437 measurement. Details for the material and experimental methods and results could be found
438 in their publication. Because the suctions of the various concentrations of NaCl were not
439 measured in their study, these suctions were determined based on a method proposed by Lang
440 (1976).

441 With the exception of sample CB-0, the difference between the total suction of a sat-
442 urated sample and the suction of the hydrating fluid used to saturate the sample was nearly
443 equal to the swelling pressure of the saturated sample for both bentonites. The results pre-
444 sented in Fig. 9 validate Eq. (9) that was derived based on the thermodynamics of soil
445 moisture. For sample CB-0, the difference in its total suction and the suction of deionized
446 water used to saturate the sample was found to be greater than its swelling pressure. As
447 explained above, the significant total suction measurement error (i.e. $\pm 140\%$) could have
448 overestimated the real total suction of sample CB-0. The results in Fig. 9 show that there
449 is a strong linkage between the swelling pressure of saturated bentonites, the total suction of
450 saturated bentonites and the suction of hydrating fluid. Measurements of any two parameters
451 would be sufficient to assess the unknown parameter. The linkage between these three param-
452 eters indicates that the total suction of a bentonite caused by the adsorptive and osmotic forces
453 would not dissipate when the bentonite is saturated with aqueous solutions under constant
454 volume condition.

455 5. Conclusions

456 The linkage between the swelling pressure of saturated bentonites, the total suction of
457 saturated bentonites and the suction of the aqueous solutions used to saturate bentonites was

458 examined based on the thermodynamics of soil moisture. To validate the derived equation,
459 constant volume swelling pressure tests and suction measurements were carried out on the Ca-
460 Mg-rich bentonite in the laboratory. The following conclusions were drawn from the study.

461 1. The time taken for the swelling pressure to reach an equilibrium increased with
462 the increase in the dry density of the bentonite, which was attributed to a decrease in the
463 permeability of the bentonite at high compaction dry densities.

464 2. In addition to the swelling pressure of the saturated bentonite, the total suction
465 of the saturated bentonite was significantly affected by the compaction dry density of the
466 bentonite and the salt concentration of the hydrating fluids, too. The increase in the total
467 suction of the saturated bentonite with the compaction dry density results from the decrease
468 the amount of water present in the saturated bentonite. The increase in the total suction of the
469 saturated bentonite with the salt concentration of the hydrating fluids is attributed primarily
470 to an increase in the osmotic suction caused by an increase in the amount of dissolved salts
471 in the macrostructural pore fluids.

472 3. The total suction of the saturated bentonite was nearly equal to the swelling pressure
473 in case of water as the hydrating fluid. However, the total suction was significantly greater
474 than the swelling pressure in case of CaCl_2 solutions as the hydrating fluid. Moreover, the total
475 suction of the saturated bentonite was found to be greater than the suction of the hydrating
476 fluids.

477 4. Based on the experimental results from this study and those reported in the literature
478 it was found that the difference between the total suction of saturated bentonites and the
479 suction of the aqueous solutions used to saturate the bentonites is approximately equal to
480 the swelling pressure of saturated bentonites. The linkage between these three parameters
481 indicates that the total suction of a bentonite caused by the adsorptive and osmotic forces
482 would not dissipate when the bentonite is saturated with aqueous solutions under constant
483 volume condition. The present finding extends the relationship suggested by Croney (1952);

484 Kassiff and Shalom (1971); Townr (1981); Karnland et al. (2005). Moreover, the linkage
485 between these three parameters provides the framework for establishing a quantitative linkage
486 between swelling pressure of bentonites and hydro-mechanical-chemical boundary conditions
487 in the deep geological repositories.

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*Highlights (for review)

- Time for swelling pressure to reach equilibrium increases with bentonite density.
- Bentonite density and concentration of solutions affect the suction of bentonites.
- Swelling pressure equals the difference in suction between bentonite and solutions.

Table 1. Initial compaction conditions of bentonite samples and the types of aqueous solution used for saturating the samples during the swelling pressure tests

Sample no.	Initial compaction conditions				Aqueous solutions
	Dry density	Water content	Degree of saturation	Total soil suction*	
	(Mg/m ³)	(%)	(%)	(MPa)	
CB-0	1.20	20.0	42.0	13.7	Deionized water
CB-1	1.41	20.0	56.2	13.7	Deionized water
CB-2	1.57	20.0	71.6	13.7	Deionized water
CB-3	1.57	20.0	71.8	13.7	Deionized water
CB-4	1.58	20.0	72.6	13.7	Deionized water
CB-5	1.66	20.0	81.6	13.7	Deionized water
CB-6	1.83	16.4 ^α	86.2	52.4	Deionized water
CB-7	1.59	9.0	33.3	166.3	Deionized water
CB-8	1.59	9.0	33.2	166.3	0.5 M CaCl ₂
CB-9	1.59	9.0	33.3	166.3	1.0 M CaCl ₂
CB-10	1.59	9.0	33.3	166.3	2.0 M CaCl ₂

*, the targeted water content of 20% could not be achieved due to experimental error; *, determined using chilled-mirror hygrometer and Kelvin's law.

Figure

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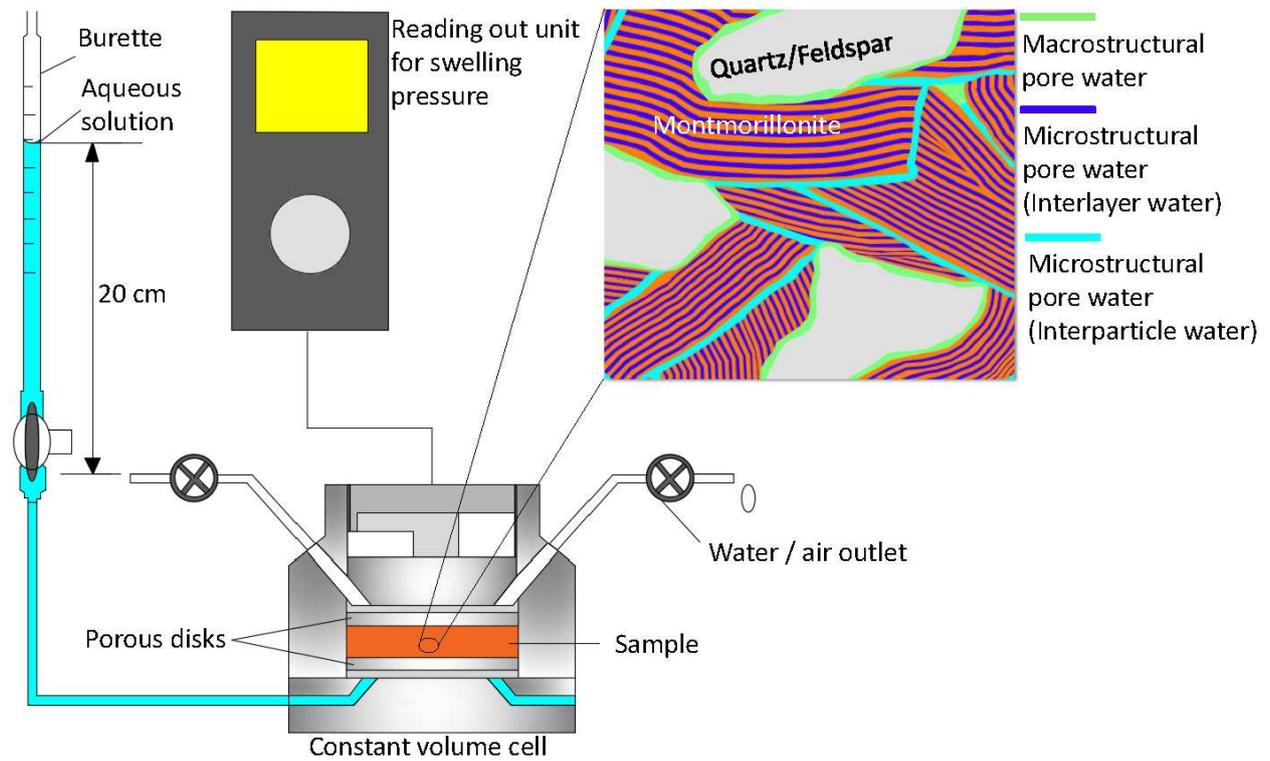


Fig. 1. Schematics of swelling pressure measurement using constant volume test set-up and of structure of compacted bentonite sample

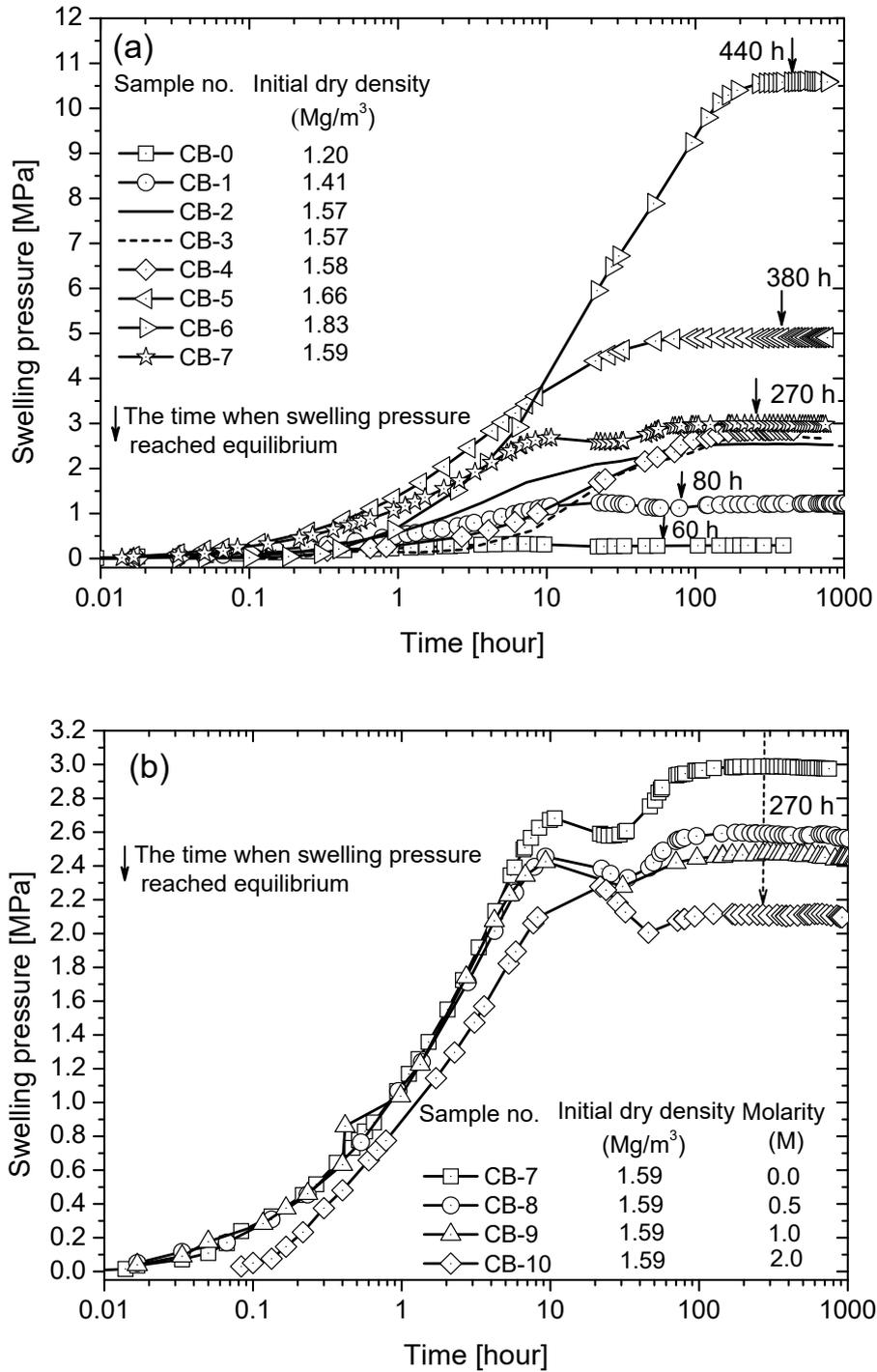


Fig. 2. Elapsed time versus swelling pressure for the samples (a) saturated with deionized water and (b) saturated with CaCl₂ solutions of different concentrations

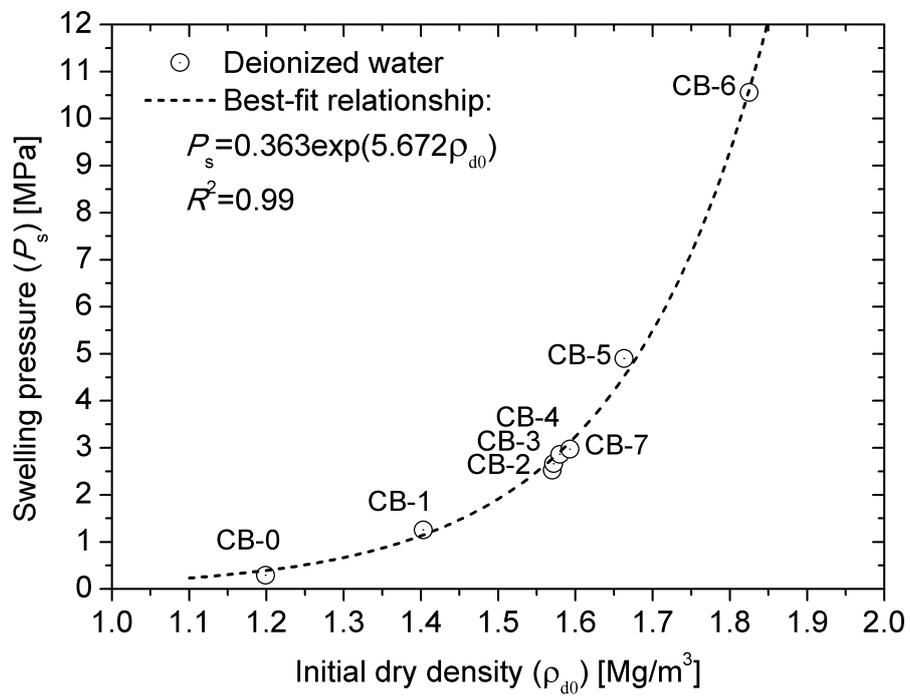


Fig. 3. Effect of initial dry density on swelling pressure

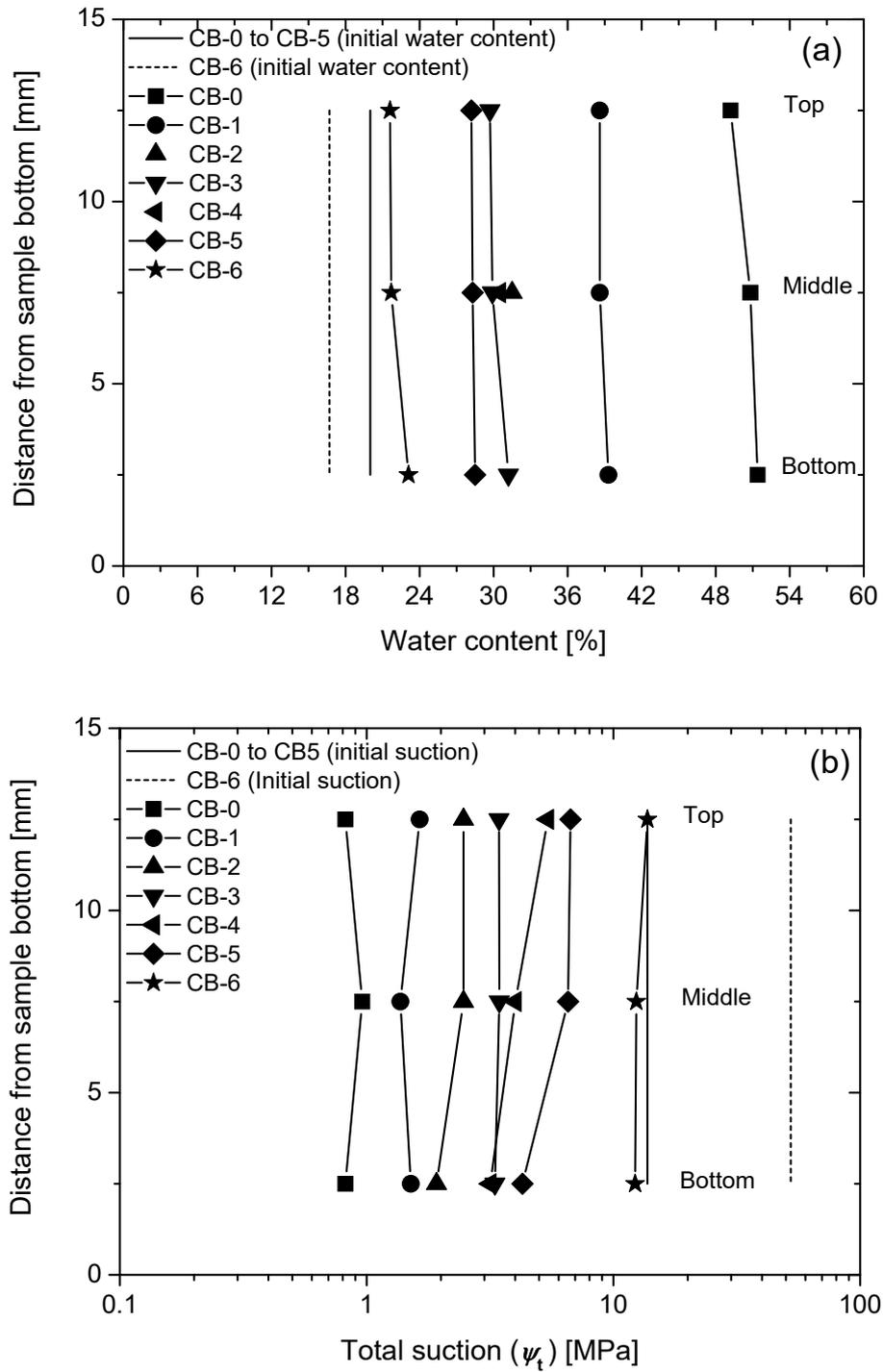


Fig. 4. Profiles of (a) water content and (b) total suction for the swelling pressure tests with deionized water (water contents of samples CB-2 and CB-4 correspond to mean water contents)

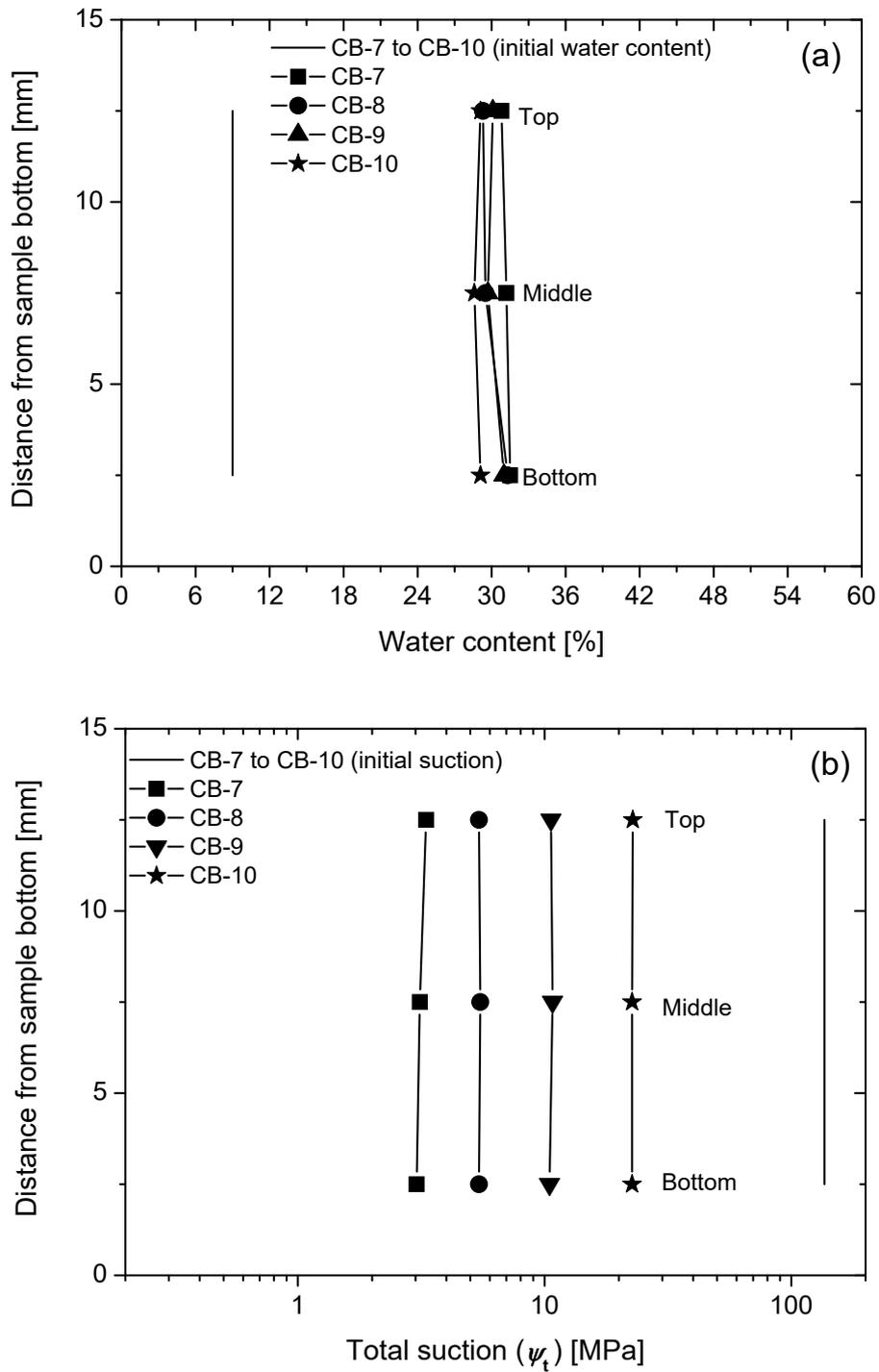


Fig. 5. Profiles of (a) water content and (b) total suction for the swelling pressure tests with CaCl_2 solutions

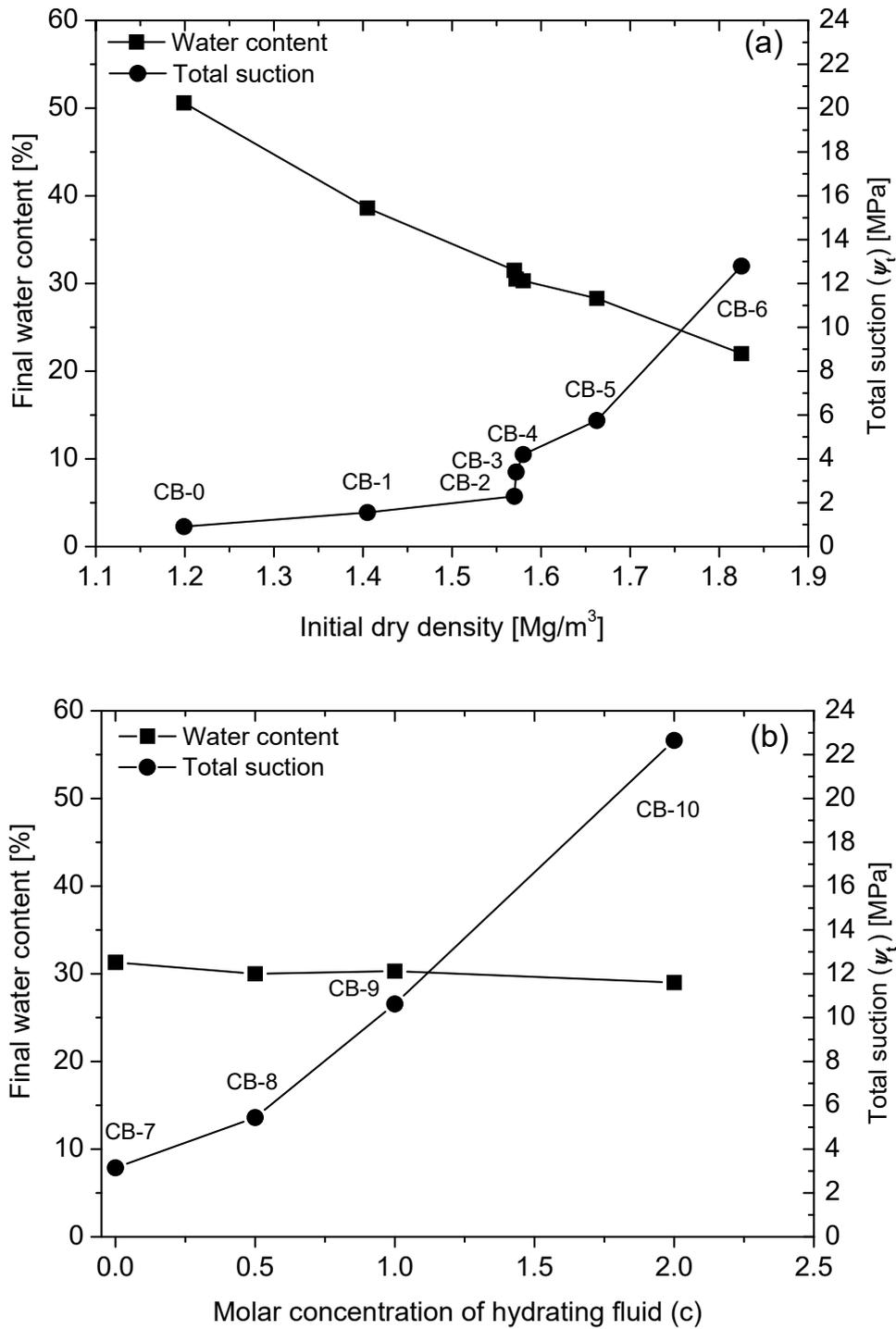


Fig. 6. Effects of (a) initial dry density and (b) CaCl_2 concentration on the water content and total suction of the saturated bentonite samples after swelling pressure tests

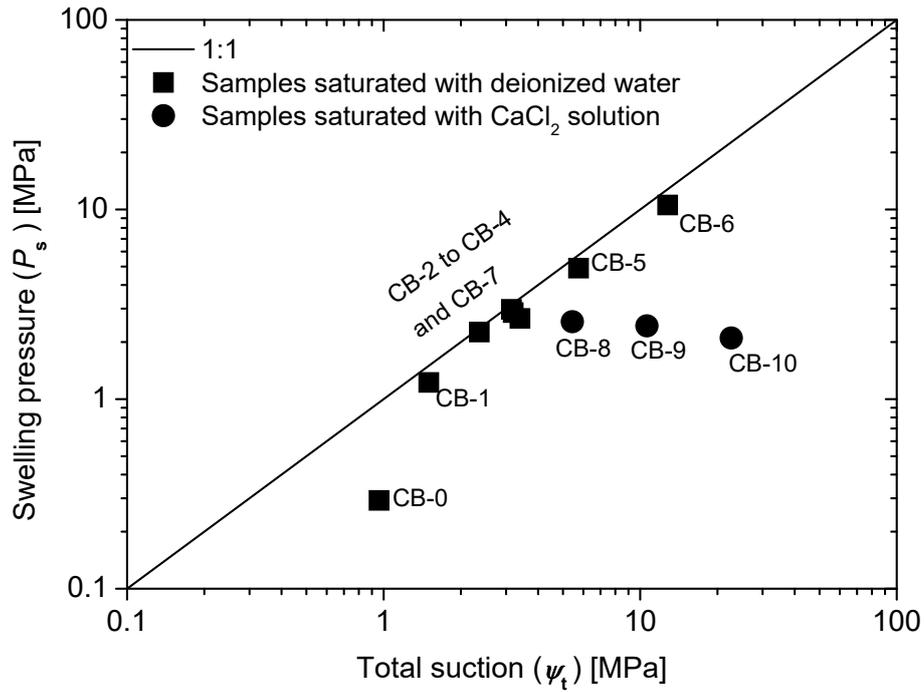


Fig. 7. Total suction versus swelling pressure plot for saturated bentonite samples at the end of swelling pressure tests

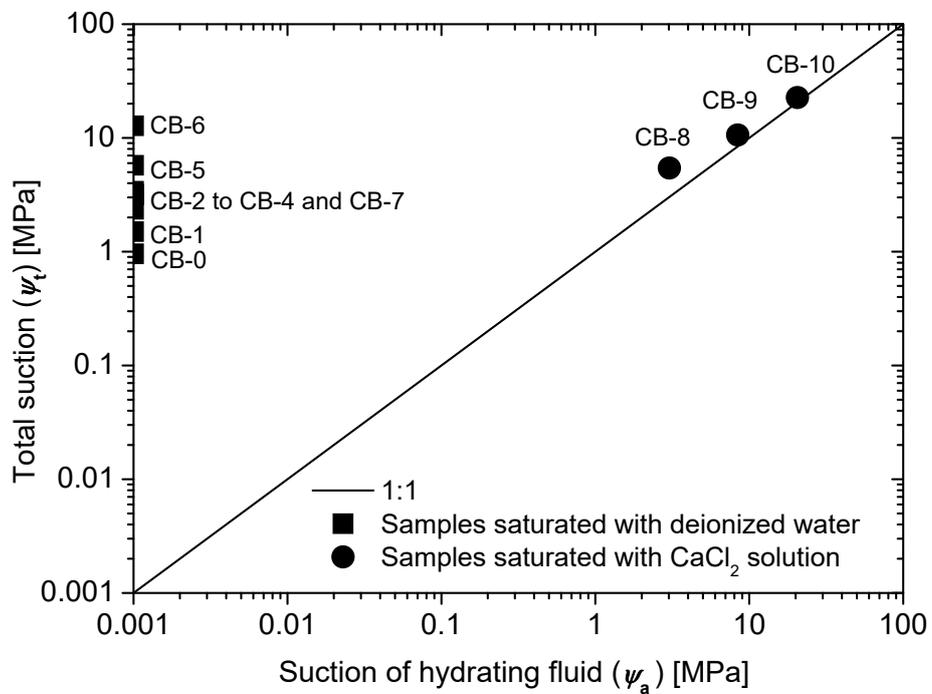


Fig. 8. Suction of hydrating fluid versus total suction of the saturated samples at the end of the swelling pressure tests

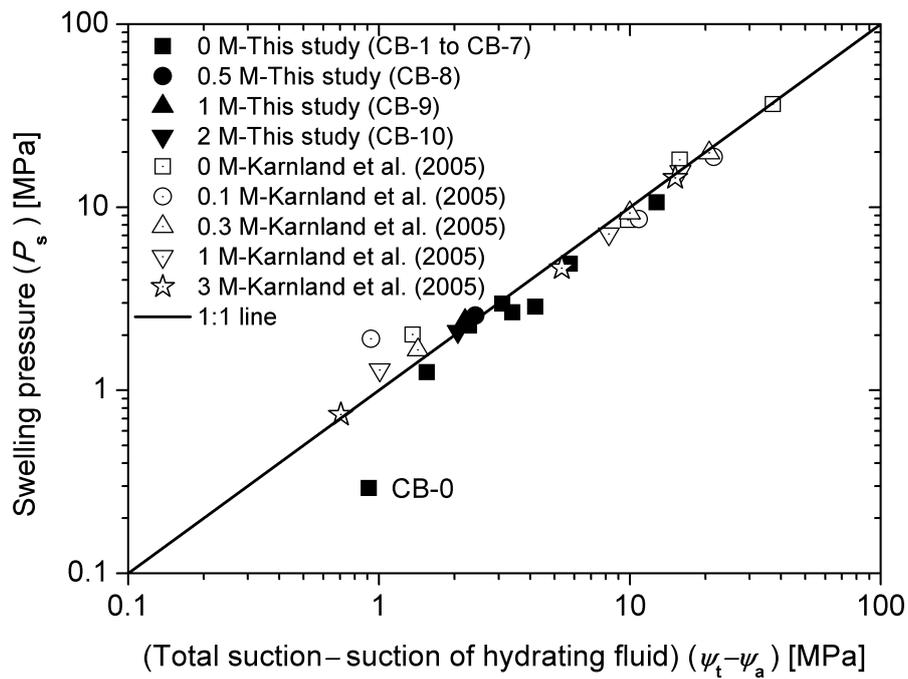


Fig. 9. Difference between the total suction of saturated bentonites and the suction of hydrating fluid versus the swelling pressure of saturated bentonites for tests with deionized water and various salt solutions (0 M in the legend corresponds to deionized water)

Linkage between swelling pressure, total suction of saturated bentonites and suction of saturating aqueous solutions

Abstract: In deep geological repositories, compacted bentonites have been proposed for use as barrier materials for isolating nuclear waste. The prevailing hydro-mechanical-chemical boundary conditions in the repositories can affect the swelling capacity of compacted bentonites. The present study examines the linkage between the swelling pressure of saturated bentonites, the total suction of saturated bentonites and the suction of hydrating fluids. An equation describing the linkage between these parameters was derived based on the thermodynamics of soil moisture. To validate the derived relationship, laboratory tests involving constant volume swelling pressure tests and total suction measurements after the swelling pressure tests were carried out on a selected Ca-Mg-rich bentonite. Seven bentonite samples with compaction dry density ranging from 1.20 and 1.83 Mg/m³ were hydrated with deionized water, whereas four samples with a dry density of 1.59 Mg/m³ were hydrated with 0.0001 to 2.0 M CaCl₂ solutions. The total suctions of the saturated bentonite and the hydrating fluids were determined by using a chilled-mirror hygrometer. The total suction of the saturated bentonite was found to be affected by the compaction dry density and the suction of the hydrating fluids. The experimental results from the present study and from the literature demonstrated that the total suction of saturated bentonites minus the suction of the hydrating fluid used to saturate bentonites is approximately equal to the swelling pressure of saturated bentonites.

Key words: Expansive soils; bentonite; swelling pressure; suction; thermodynamics of soil moisture; radioactive waste disposal

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