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

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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/m^3 were hydrated with deionized water, whereas four samples with a dry density of 1.59 Mg/m^3 were hydrated with 0.0001 to 2.0 M $CaCl_2$ 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.

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

1. Introduction

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

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

The total suction of unsaturated soils consists of matric and osmotic suctions (Yong and Warkentin, 1975; Fredlund et al., 2012; Lu and Likos, 2004). In clays, the matric suction is due to surface tension, adsorptive forces and osmotic forces (i.e. the diffuse double layer forces), whereas the osmotic suction is due to the presence of dissolved solutes in the pore water.

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

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

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

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

The objective of this study was to examine the linkage between the swelling pressure, the total suction and the suction of hydrating fluids in case of saturated bentonites. An equation describing the linkage between these three parameters was first derived based on the thermodynamics of soil moisture. Further, constant volume swelling pressure tests and total suction measurements after the swelling pressure tests were carried out on a selected bentonite. Based on the experimental results from this study and from the literature, the derived equation was then validated.

2. Theoretical considerations

Total potential of soil-water has been discussed from the point of view of thermodynamic considerations of soil moisture by numerous researchers (Edlefsen and Anderson, 1943; Low and Deming, 1953; Bolt and Miller, 1958; Mitchell, 1962; Yong and Warkentin, 1975; Iwata et al., 1995). Iwata et al. (1995) stated that the total water potential (μ) in clays relative to the potential of the free pure water at the same temperature and external pressure is comprised of the potentials due to surface tension (μ_{c}), van der Waals force (μ_{f}), solutes (μ_0), electric field (μ_{e}), temperature (μ_{T}), pressures (μ_{P}), and gravitational field (μ_{g}) (Eq. 1).

$$\mu = \mu_{\mathrm{c}} + \mu_{\mathrm{f}} + \mu_0 + \mu_{\mathrm{e}} + \mu_{\mathrm{T}} + \mu_{\mathrm{P}} + \mu_{\mathrm{g}} \tag{1}$$

Detailed description about these potentials could be found in the work of Iwata et al. (1995). Since μ_{c}, μ_{f}, μ_0, μ_{e} and μ_{P} are the potentials which are closely related the discussion in this paper, their definitions are given in the following paragraphs.

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

Van der Waals force effect (μ_{f}) corresponds to the potential energy of interaction between clay surface and a water molecule. Therefore, μ_{f} results in a decrease in the total potential of the water and the decrease in the total potential of soil water depends upon the distance from the surface of the clay particle to a water molecule.

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

Clay minerals have electric charge and the soil solution is affected by the electric field induced by those charges. The change of the total potential of water in soil because of electric displacement is defined as μ_{e}.

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

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

Sketches of a swelling pressure test device and of the structure of compacted bentonite (macro and micro structure) are shown in Fig. 1. A compacted bentonite sample under constant volume condition is hydrated with an aqueous solution. The upon saturation of the sample in the measuring system (Fig. 1) during a swelling pressure test, the macrostructural pores are filled with the aqueous solution. In addition, the microstructural pores are also filled with water that may also include some solutes. At saturation, the water potential caused by surface tension (μ_{c}) gets eliminated. Considering that the temperature and the atmospheric pressure change little during an isothermal swelling pressure test, the potentials caused by temperature (μ_{T}) and atmospheric pressure can be neglected. Since the scale of this system is very small, it is reasonable to neglect the gravitational potential (μ_{g}). Therefore, the total potential of the macrostructural pore water (μ_{M}) would be reduced to only the potential due to solutes (μ_0^{M}) and is given by Eq. (2). In a swelling pressure test, μ_0^{M} corresponds to the total water potential of the hydrating fluid (μ_{a}). In suction terms, the total suction of the hydrating fluid is ψ_{a}. In Eq. (2), $\overline{\upsilon}_{\mathrm{w}}$ is the partial specific volume of water.

$$\mu_{\mathrm{M}} = \mu_{\mathrm{a}} = -\psi_{\mathrm{a}}\overline{\upsilon}_{\mathrm{w}} \tag{2}$$

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

$$\mu_{\mathrm{m}} = \mu_{\mathrm{f}}^{\mathrm{m}} + \mu_0^{\mathrm{m}} + \mu_{\mathrm{e}}^{\mathrm{m}} + P\overline{\upsilon}_{\mathrm{w}} \tag{3}$$

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

$$\mu_{\mathrm{m}} = \mu_{\mathrm{M}} \tag{4}$$

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

$$\mu_{\mathrm{f}}^{\mathrm{m}} + \mu_{\mathrm{e}}^{\mathrm{m}} + \mu_0^{\mathrm{m}} = -\psi_{\mathrm{a}}\overline{\upsilon}_{\mathrm{w}} - P\overline{\upsilon}_{\mathrm{w}} \tag{5}$$

It is generally assumed that $\overline{\upsilon}_{\mathrm{w}}$ does not depend on pressure and is a constant, nearly 1 m^3/Mg. A simplification of Eq. (5) yields Eq. (6).

$$-\frac{1}{\overline{\upsilon}_{\mathrm{w}}}(\mu_{\mathrm{f}}^{\mathrm{m}} + \mu_{\mathrm{e}}^{\mathrm{m}} + \mu_0^{\mathrm{m}}) - \psi_{\mathrm{a}} = P \tag{6}$$

Since $(\mu_{\mathrm{f}}^{\mathrm{m}} + \mu_{\mathrm{e}}^{\mathrm{m}})$ and μ_0^{m} correspond to the hydration effect and osmotic effect, respectively, thus they correspond to matric potential and osmotic potential, respectively. Because $(\mu_{\mathrm{f}}^{\mathrm{m}} + \mu_{\mathrm{e}}^{\mathrm{m}} + \mu_0^{\mathrm{m}})$ corresponds to the sum of matric and osmotic potentials in case of saturated clays, their value can be approximately determined from Kelvin's equation (i.e. Eq. (7)).

$$\mu_{\mathrm{f}}^{\mathrm{m}} + \mu_{\mathrm{e}}^{\mathrm{m}} + \mu_0^{\mathrm{m}} = \frac{RT}{M_{\mathrm{w}}}\ln(RH) \tag{7}$$

In Eq. (7), R is the molar gas constant, M_{w} is the molecular mass of water, and RH is the relative humidity. In suction terms, the total suction (ψ_{t}) of the saturated bentonite under constant volume condition is given in Eq. (8)

$$\psi_{\mathrm{t}} = -\frac{RT}{M_{\mathrm{w}}\bar{v}_{\mathrm{w}}}\ln(RH) \tag{8}$$

After Eqs. (6), (7), and (8) are combined, Eq. (9) can be obtained in suction and pressure terms, where P_{s} represents swelling pressure.

$$\psi_{\mathrm{t}} - \psi_{\mathrm{a}} = P = P_{\mathrm{s}} \tag{9}$$

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

$$s + U = P \tag{10}$$

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

To validate Eq. (9) experimentally, separate measurements of P_{s}, ψ_{a} and ψ_{t} are necessary. The former two can be directly determined by a swelling pressure device and a chilled-mirror hygrometer, respectively. However, the latter cannot be directly determined by placing a relative humidity sensor in a constant volume swelling pressure device. In constant volume condition, the water potential determined by the relative humidity sensor is the total water potential (μ_{m} or μ_{M}). This can be confirmed by constant volume infiltration experimental results reported by Cui et al. (2008); Ye et al. (2009); Mokni and Barnichon (2016); Villar et al. (2016). They found that the relative humidity above the wetting face reached nearly 100% after long-term of supplying water. This indicates that the water potential determined by the relative humidity sensor in constant volume condition is equal to the water potential of the hydrating fluid (water). Thus, releasing swelling pressure by removing the confinement conditions is necessary for determining ψ_{t} or $(\mu_{\mathrm{f}}^{\mathrm{m}} + \mu_{\mathrm{e}}^{\mathrm{m}} + \mu_{0}^{\mathrm{m}})$.

If the hydrating fluid (Fig. 1) is removed before releasing the swelling pressure, subsequent removal of swelling pressure can be considered as undrained unloading. Consequently, the water content of the saturated bentonite sample would not change after releasing the swelling pressure. Since $\mu_{\mathrm{f}}^{\mathrm{m}}$ and $\mu_{\mathrm{e}}^{\mathrm{m}}$ are dominated by water content or volume, it is reasonable to assume that they would not change after releasing the swelling pressure. Because μ_{0}^{m} is dominated by the molarity of solutes and the unloading does not change the mass of solutes and the water content, μ_{0}^{m} would remain after releasing the swelling pressure. This would be also true for μ_{0}^{M}. Thus, $(\mu_{\mathrm{f}}^{\mathrm{m}} + \mu_{\mathrm{e}}^{\mathrm{m}} + \mu_{0}^{\mathrm{m}})$ or ψ_{t} remains unchanged before and after releasing the swelling pressure.

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

$$\mu_{\mathrm{M}}^{\mathrm{un}} = \mu_{0}^{\mathrm{M}} - (u_{\mathrm{a}} - u_{\mathrm{w}})\overline{v}_{\mathrm{w}} = \mu_{0}^{\mathrm{M}} - P_{\mathrm{s}}\overline{v}_{\mathrm{w}} = -(\psi_{\mathrm{a}} + P_{\mathrm{s}})\overline{v}_{\mathrm{w}} \tag{11}$$

Because of the removal of the swelling pressure, the total potential of the microstructural pore water ($\mu_{\mathrm{m}}^{\mathrm{un}}$) would be reduced to Eq. (12).

$$\mu_{\mathrm{m}}^{\mathrm{un}} = \mu_{\mathrm{f}}^{\mathrm{m}} + \mu_{\mathrm{e}}^{\mathrm{m}} + \mu_{0}^{\mathrm{m}} = -\psi_{\mathrm{t}}\overline{v}_{\mathrm{w}} \tag{12}$$

Comparison of Eq. (11) and Eq. (12) indicates that the total potential of the macrostructural pore water would be still equal to the total potential of the microstructural pore water after releasing the swelling pressure. In other words, the thermodynamic equilibrium remains during undrained unloading, which would mean that there is no water exchange between the macrostructural pore and the microstructural pore water or that the assumption of undrained unloading is reasonable. Thus, after releasing the swelling pressure, the total suction of the saturated bentonite would be equal not only to the capillary suction triggered by swelling pressure release but also to the total suction (ψ_{t}) of the saturated bentonite under constant volume condition. Consequently, ($\mu_{\mathrm{f}}^{\mathrm{m}} + \mu_{\mathrm{e}}^{\mathrm{m}} + \mu_{0}^{\mathrm{m}}$) or ψ_{t} can be also determined by a chilled-mirror hygrometer after releasing the swelling pressure.

To validate Eq. (9) experimentally, constant volume swelling pressure tests were performed on a Ca-Mg-rich bentonite using various aqueous solutions, i.e. deionized water and solutions of $CaCl_2$. After the swelling pressure tests, the suctions (ψ_{a}) of the saturating aqueous solutions and the total suctions (ψ_{t}) of the saturated bentonite samples were determined.

3. Materials and methods

3.1. Materials

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

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

3.2. Experimental methods

3.2.1. Sample preparation

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

Compacted bentonite samples, 50 mm in diameter and 15±0.02 mm in height, were prepared by statically compacting bentonite-water mixtures inside the oedometer ring by using a 15-tonne capacity compression testing machine. The samples were compacted by applying uniaxial loads. The initial compaction conditions of the as-compacted samples and aqueous solutions used to saturate the samples during the swelling pressure tests are shown in Table 1.

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

An isochoric swelling pressure device (Fig. 1) (Romero, 1999; Schanz and Tripathy, 2009) was used in this study for measuring the swelling pressures of compacted bentonite samples. After the static compaction process, the as-compacted sample inside the sample ring was placed in the swelling pressure device. Samples CB-0 to CB-6 with compaction dry densities varying between 1.20 to 1.83 Mg/m^3 (Table 1) were hydrated by supplying deionized water. Samples CB-2, CB-3 and CB-4 had very similar compaction conditions (dry density = 1.57 Mg/m^3 and water content = 20%). Samples CB-7, CB-8, CB-9, and CB-10 (dry density = 1.59 Mg/m^3) were hydrated with deionized water, 0.5, 1.0 and 2.0M $CaCl_2$ solutions, respectively. The hydrating fluids were supplied from burettes attached to the bottom fluid reservoirs of the devices. During the swelling pressure tests, the $CaCl_2$ solutions were renewed periodically for maintaining the constant concentration of the solutions.

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

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

3.2.3. Determination of suctions of hydrating fluids/aqueous solutions

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

$$\psi_\mathrm{a} = 38.54EC^{1.0489}. \tag{13}$$

The electrical conductivity (EC) of the deionized water used in this study was found to be about 0.03 mS/cm. Thus, the calculated suction of the deionized water used was approximately 0.001 MPa and was nearly equivalent to the suction of a 0.0001 M $CaCl_2$ solution based on van't Hoff equation. The suctions of $CaCl_2$ solutions after the swelling pressure tests were determined by using the chilled-mirror hygrometer and Kelvin's law. The suctions of 0.5 M, 1 M and 2 M $CaCl_2$ solutions were found to be 3.03, 8.42, and 20.55 MPa, respectively. These values were found to be similar to the suctions of the solutions prior to the swelling pressure tests and remained within the measurement accuracy of the chilled-mirror hygrometer.

4. Results and discussion

4.1. Swelling pressure

The elapsed time versus swelling pressure for bentonite samples CB-0 to CB-7 is shown in Fig. 2a. Similarly, Fig. 2b presents the evolution of swelling pressures in case of samples CB-7, CB-8, CB-9, and CB-10 that were hydrated with deionized water, 0.5, 1.0 and 2.0 M $CaCl_2$ solutions, respectively. Swelling pressure equilibrium time was defined as the time when the variation of swelling pressure started to be less than 5 kPa per day. The swelling pressure equilibrium time was found to increase with an increase in the sample dry density (Fig. 2a). The equilibrium time for sample CB-0 (dry density = 1.2 Mg/m^3) was about 60 h, whereas it was found to be about 440 h for sample CB-6 with a compaction dry density of 1.83 Mg/m^3. For a given dry density of 1.59 Mg/m^3, the type of hydrating fluid (i.e., water or $CaCl_2$ solutions) had no significant influence on the equilibrium time. In this case, the swelling pressure equilibrated in about 270 h.

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

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

In the case of samples with a constant dry density of 1.59 Mg/m^3, the equilibrium swelling pressure decreased with an increase in $CaCl_2$ concentration (Fig. 2b). A reduction in the swelling pressure from 2.98 to 2.10 MPa was found to occur (i.e. a decrease of about 30%) when the concentration of $CaCl_2$ was increased from 0.0001 to 2.0 M. Similar experimental results concerning the influence of salt solution concentration on the swelling pressure have been reported by several researchers (Pusch, 1980*b*; Bucher and Max, 1989; Pusch et al., 1990; Karnland et al., 2005; Villar and Lloret, 2008*a*; Castellanos et al., 2008; Zhu et al., 2013; Tripathy et al., 2014b).

4.2. Water content and total suction profiles

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

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

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

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

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

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

4.3. Total suction and swelling pressure

The experimental results presented in Figs. 3 and 6 show that both swelling pressure and total soil suction were dependent on the dry density of the bentonite and salt concentration of the hydrating fluids. Total suction versus swelling pressure plot is shown in Fig. 7. For the bentonite samples that were hydrated with deionized water (samples CB-0 to CB-7), with the exception of sample CB-0, the total suction was nearly equal to the swelling pressure. For sample CB-0, the total suction (0.8 MPa) was greater than the swelling pressure (0.3 MPa), which was probably the result of a significant total suction measurement error. As mentioned in the section of Materials and methods, the total suction measurement error was up to ±140% if the total suction of a soil was less than 0.3 MPa. This significant error (i.e. ±140%) could have overestimated the real total suction of sample CB-0. In the case of the samples that were hydrated with $CaCl_2$ solutions (samples CB-8 to CB-10), the total suction was considerably greater than the corresponding swelling pressure. The test results showed that if the hydrating fluid was water ($\psi_\mathrm{a} = 0.001$ MPa), there was a good agreement between swelling pressure and total suction. Disagreement between total suction and swelling pressure was only observed for the samples that were hydrated with solutions having high salt concentration.

4.4. Suction of hydrating fluids and total suction

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

4.5. Swelling pressure and difference between total suction and suction of hydrating fluids

The swelling pressures (P_s) of saturated bentonites are plotted against the differences between the total suctions of saturated bentonites and the suctions of the hydrating fluids ($\psi_\mathrm{t} - \psi_\mathrm{a}$) in Fig. 9. In addition to the test results obtained from this study on the Ca-Mg-rich bentonite, the test results reported by Karnland et al. (2005) on a Na-bentonite are also shown in Fig. 9 for the tests with water and various NaCl solutions. In their study, the compacted Na bentonite samples under constant volume condition were saturated with solutions of concentration from 0.0001 to 3 M NaCl. After the swelling pressures reached equilibrium, the samples were extracted from the constant volume device for total suction measurement. Details for the material and experimental methods and results could be found in their publication. Because the suctions of the various concentrations of NaCl were not measured in their study, these suctions were determined based on a method proposed by Lang (1976).

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

5. Conclusions

The linkage between the swelling pressure of saturated bentonites, the total suction of saturated bentonites and the suction of the aqueous solutions used to saturate bentonites was examined based on the thermodynamics of soil moisture. To validate the derived equation, constant volume swelling pressure tests and suction measurements were carried out on the Ca-Mg-rich bentonite in the laboratory. The following conclusions were drawn from the study.

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

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

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

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

Kassiff and Shalom (1971); Towenr (1981); Karnland et al. (2005). Moreover, the linkage between these three parameters provides the framework for establishing a quantitative linkage between swelling pressure of bentonites and hydro-mechanical-chemical boundary conditions 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 (Mg/m^3)	Water content (%)	Degree of saturation (%)	Total soil suction$\star$ (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 achieved due to experimental error; $\star$, determined using chilled-mirror hygrometer and Kelvin's law.

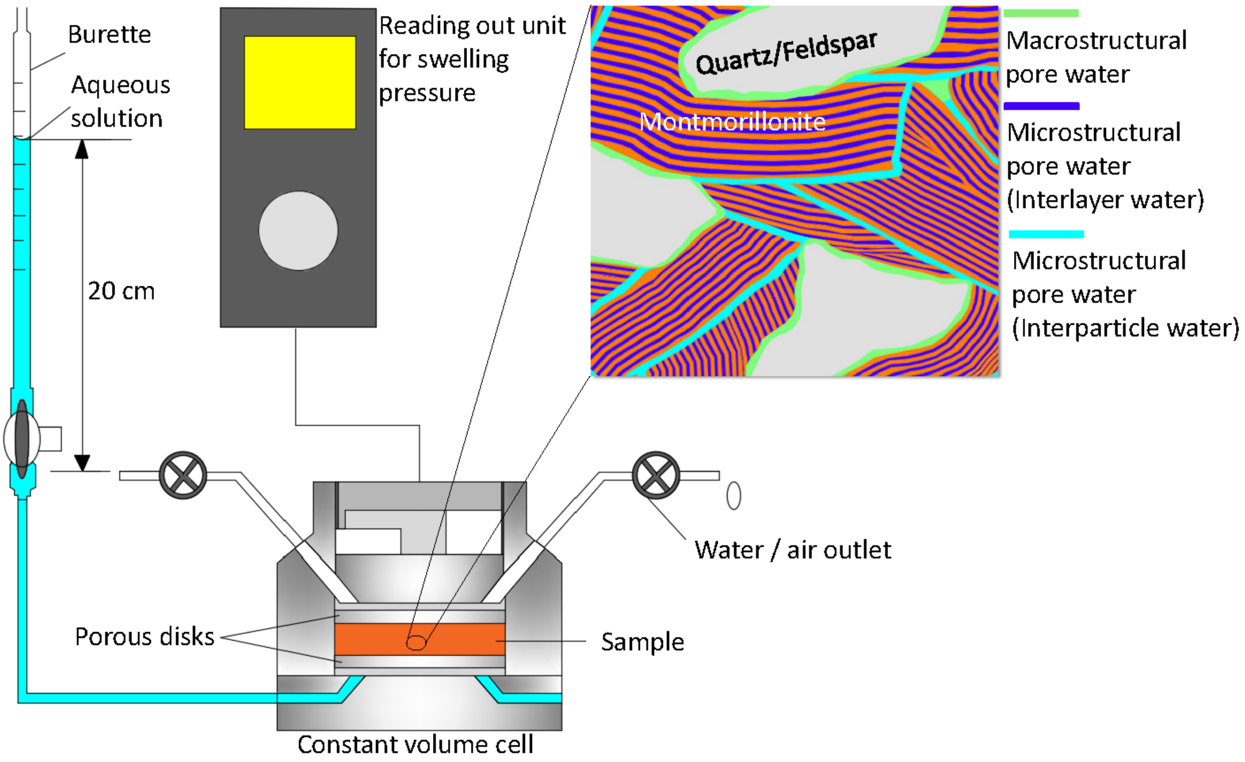

Fig. 1. Schematics of swelling pressure measurement using constant volume test set-up and of structure of compacted bentonite sample

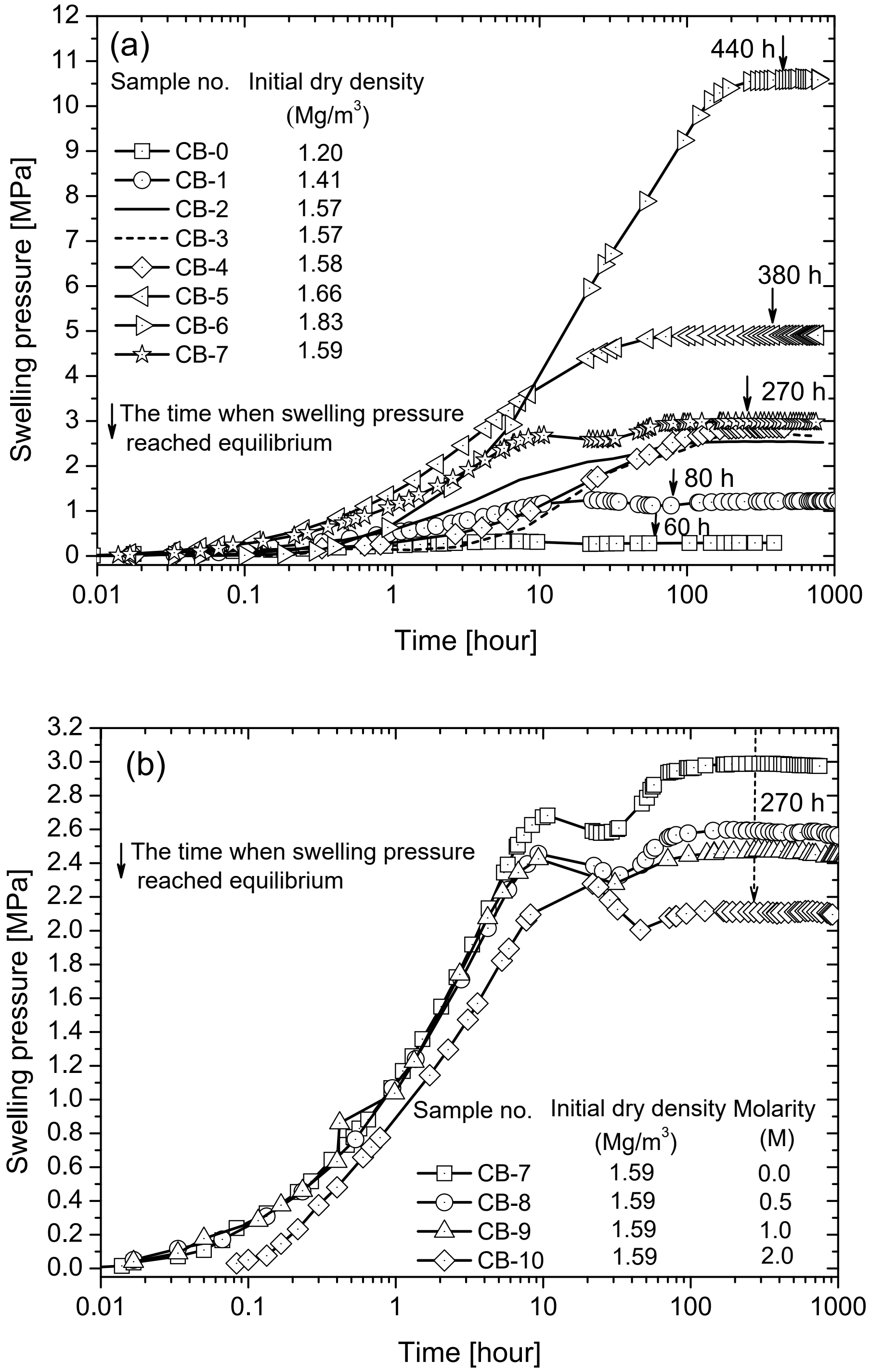

Fig. 2. Elapsed time versus swelling pressure for the samples (a) saturated with deionized water and (b) saturated with $CaCl_2$ solutions of different concentrations

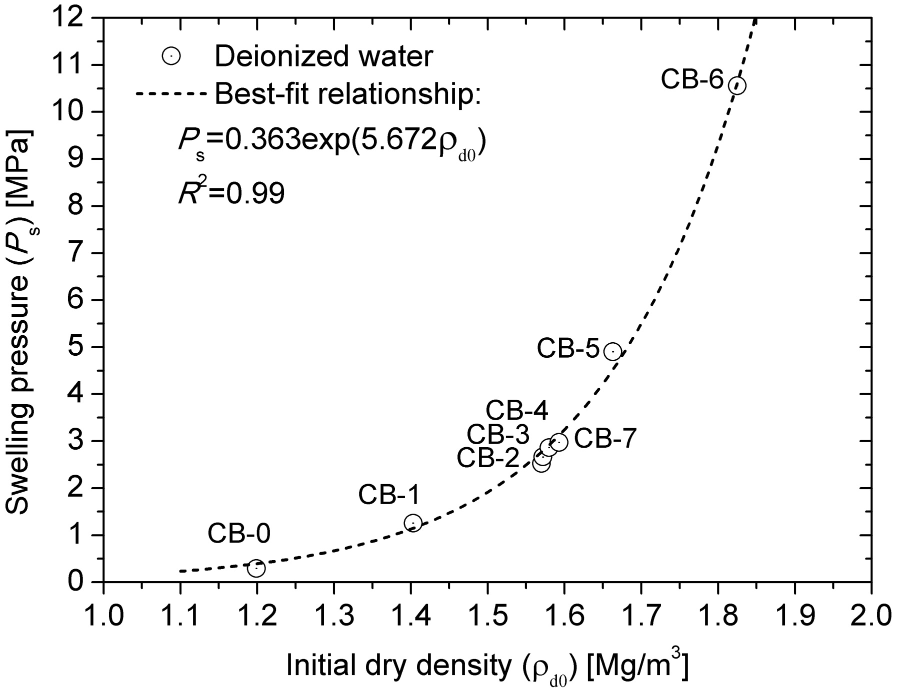

Fig. 3. Effect of initial dry density on swelling pressure

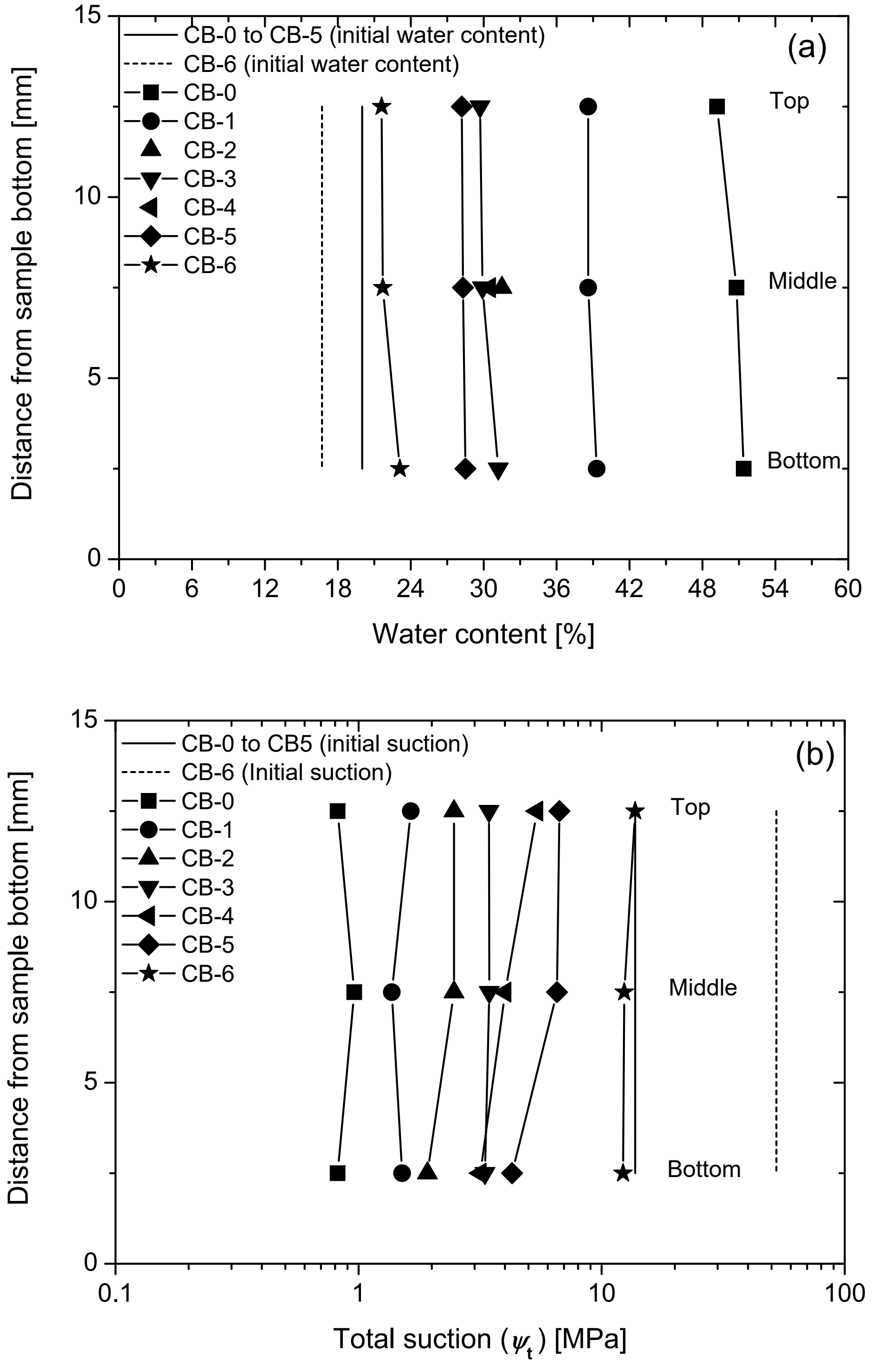

Fig. 4. Profiles of (a) water content and (b) total suction for the swelling pressure tests with deionized water (water contents of samples CB-2 and CB-4 correspond to mean water contents)

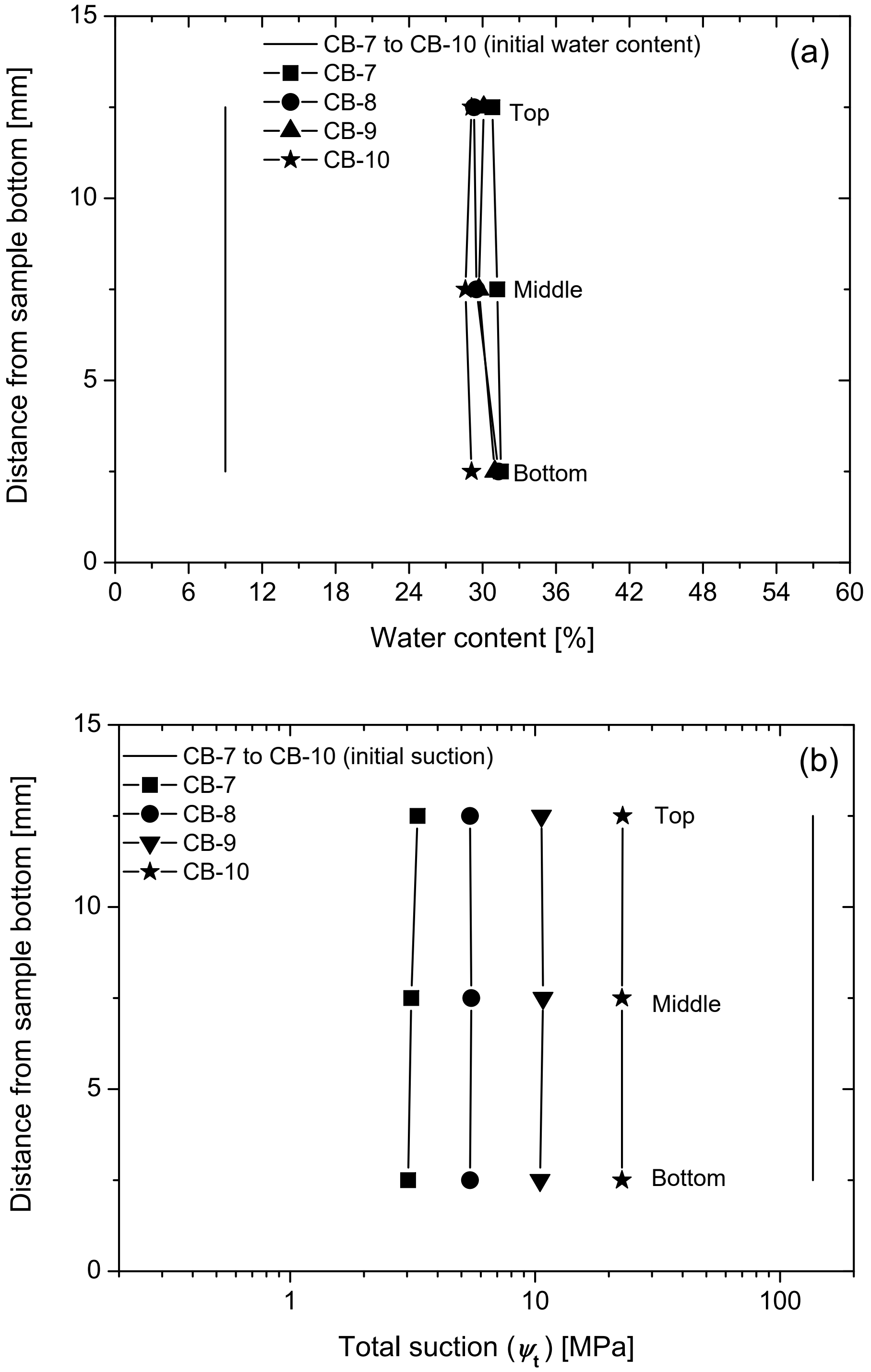

Fig. 5. Profiles of (a) water content and (b) total suction for the swelling pressure tests with $CaCl_2$ solutions

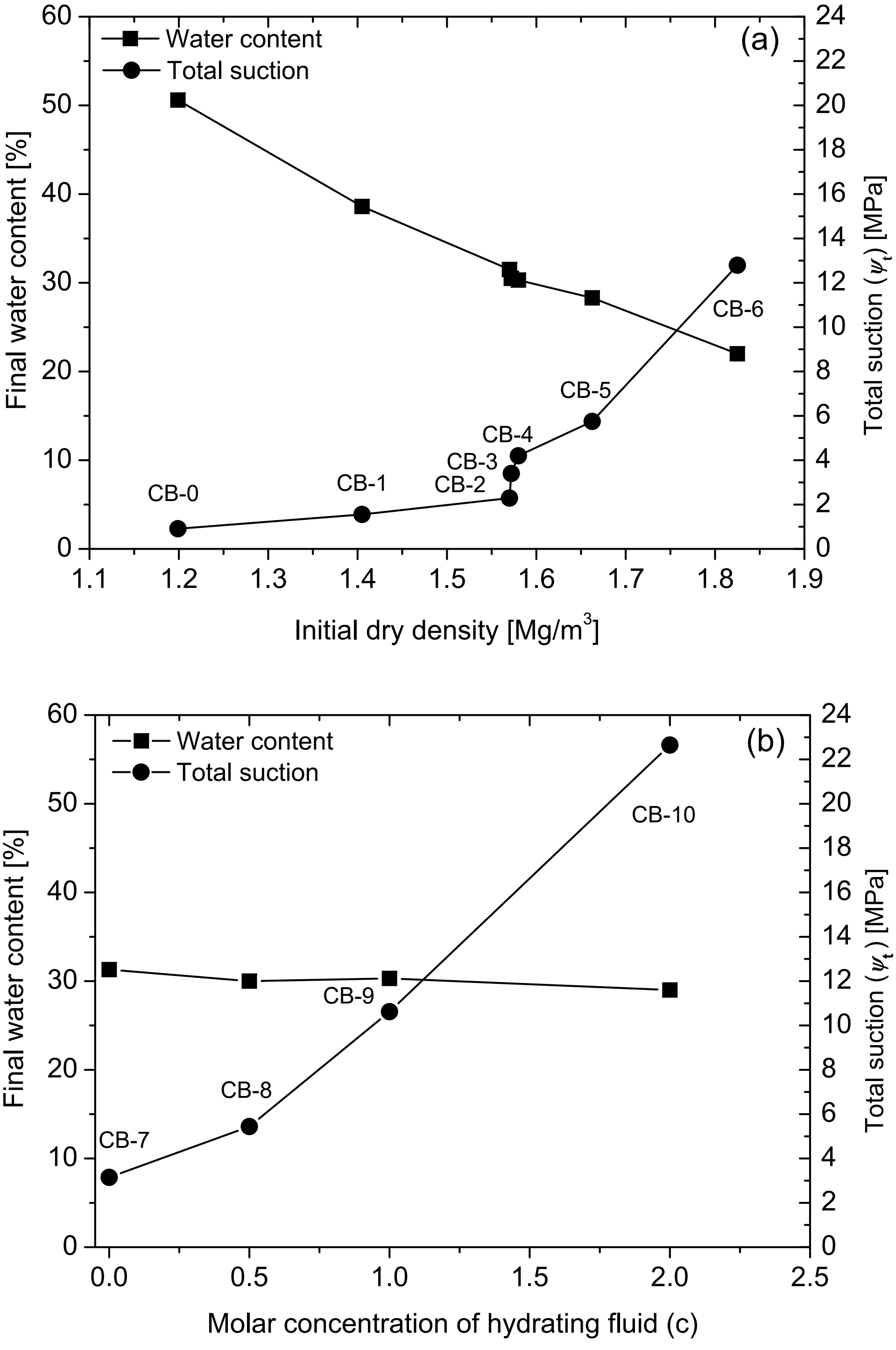

Fig. 6. Effects of (a) initial dry density and (b) $CaCl_2$ concentration on the water content and total suction of the saturated bentonite samples after swelling pressure tests

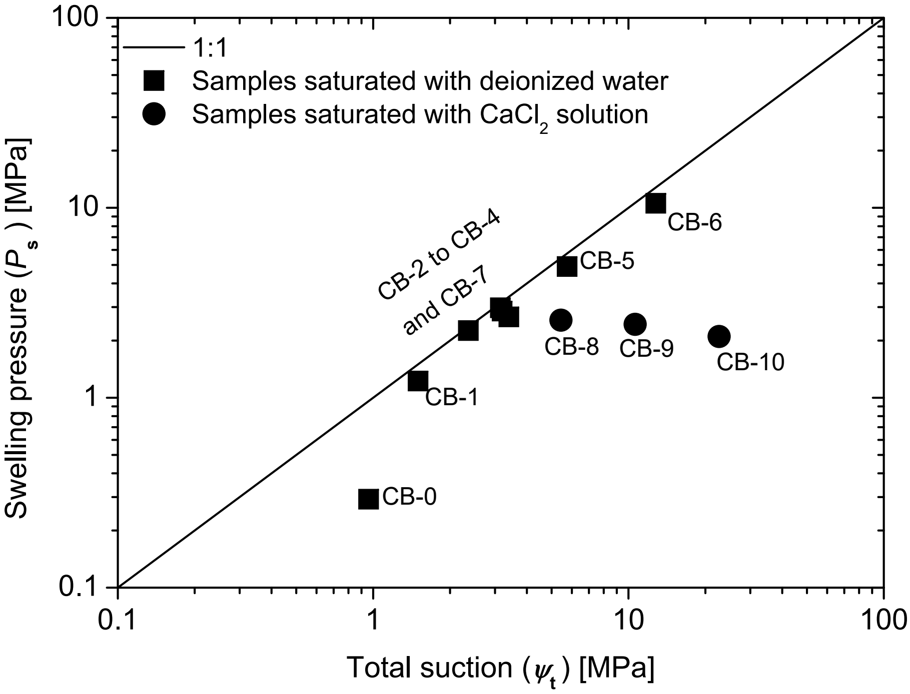

Fig. 7. Total suction versus swelling pressure plot for saturated bentonite samples at the end of swelling pressure tests

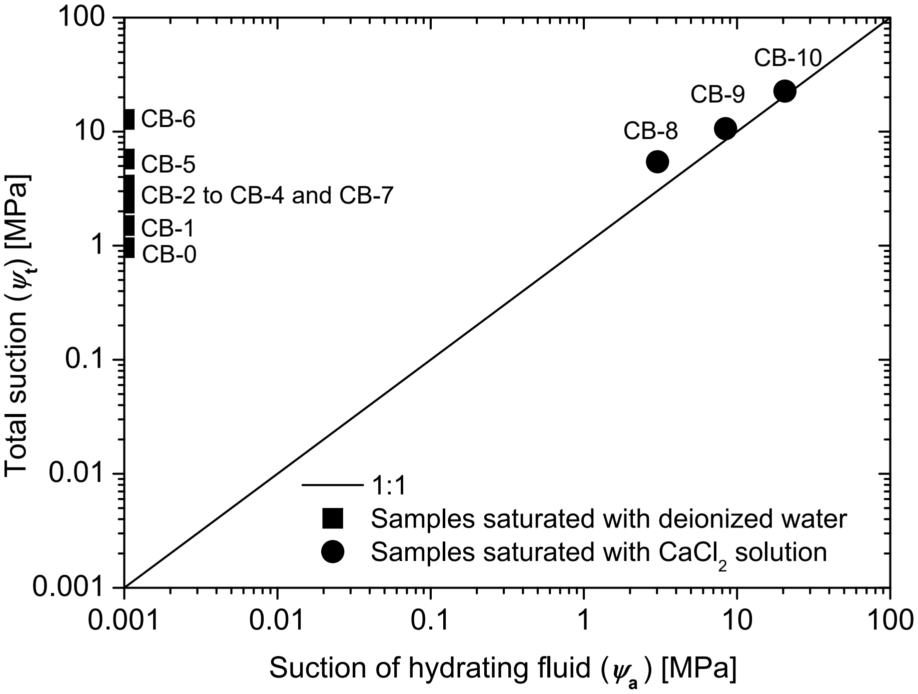

Fig. 8. Suction of hydrating fluid versus total suction of the saturated samples at the end of the swelling pressure tests

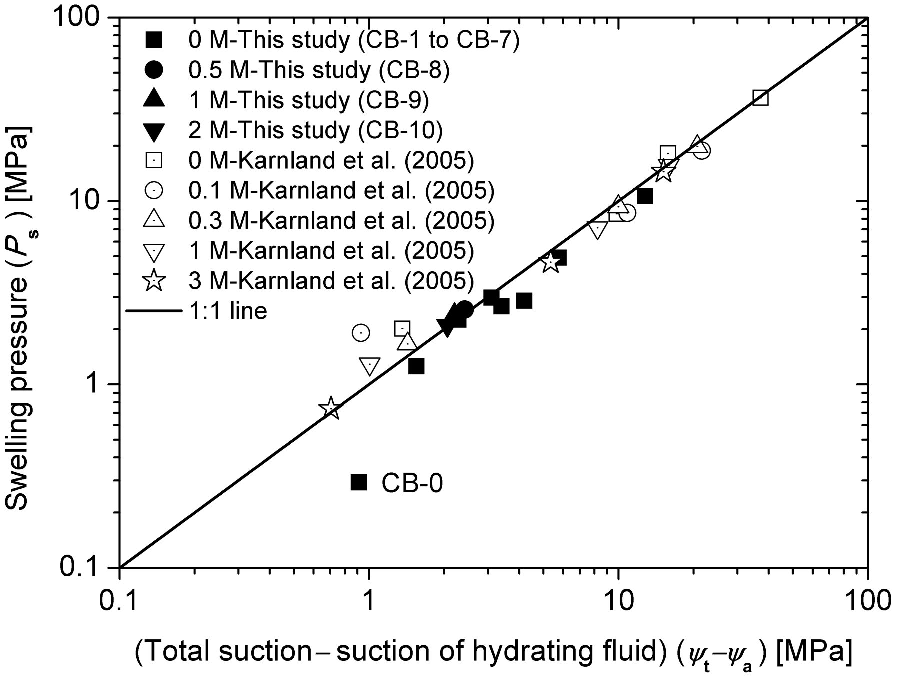

Fig. 9. Difference between the total suction of saturated bentonites and the suction of hydrating fluid versus the swelling pressure of saturated bentonites for tests with deionized water and various salt solutions (0 M in the legend corresponds to deionized water)

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/m^3 were hydrated with deionized water, whereas four samples with a dry density of 1.59 Mg/m^3 were hydrated with 0.0001 to 2.0 M $CaCl_2$ 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