# Movement of water in compacted bentonite and its relation with swelling pressure

| Journal: | Canadian Geotechnical Journal |
| --- | --- |
| Manuscript ID | cgj-2019-0219.R1 |
| Manuscript Type: | Article |
| Date Submitted by the Author: | 16-Jun-2019 |
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| Keyword: | Bentonite, swelling pressure, XRD, diffusivity, multi-ring mold |
| Is the invited manuscript for consideration in a Special Issue? | Not applicable (regular submission) |

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Movement of water in compacted bentonite and its relation with swelling pressure

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Abstract

A testing procedure was proposed to study movement water in compacted bentonite and the development of swelling pressure \((p_s)\) when compacted bentonite specimens were wetted. In this procedure, a multi-ring mold was introduced for \(p_s\) measurements, after which the specimen was sliced for X-ray diffraction test to find movement of water in the interlayer space of montmorillonite. Results revealed a relation between four phases of \(p_s\) development and evolution of four states of interlayer water molecule arrangement of montmorillonite \((L)\): when \(p_s\) reached its first peak in phase I, \(L\) moved from 1 row water arrangement \((1w)\) to at least 2w; when \(p_s\) decreased and re-increased in phase II or III, \(L\) moved from 2w to at least 3w; and when \(p_s\) reaches a steady state in phase IV for \(L = 3w\). The \(w\) distribution in the compacted bentonite was also measured as water absorption time increased. Based on those results, the global water movement was estimated in terms of diffusivity \((D)\) following a method employing Boltzmann transform. Results of comparisons implied that \(D\) calculated using this method well matched experimental data well and the method was rather easily handled.

Keywords: Bentonite, swelling pressure, XRD, diffusivity, multi-ring mold
Nomenclature

\(\chi\): Boltzmann transform \(=z/\sqrt{t}\)

\(\lambda\): wavelength of incident wave \(=1.5418 \text{ Å for Cu}\alpha\)

\(2\theta\): angle between incident and reflected waves in XRD test

\(\theta_{\text{peak}}\): \(\theta\) at peak maximum

\(\Theta\): volumetric water content.

\(\Theta_i\) and \(\Theta_s\): \(\Theta\) at initial and saturated conditions, respectively

\(\Theta_z\): \(\Theta\) at position \(z\)

\(\rho_d\): dry density of the compacted specimens

\(\rho_{\text{ini}}\) and \(\rho_{\text{fini}}\): \(\rho_d\) before and after water absorption, respectively

\(\rho_{\text{XRD}}\): \(\rho_d\) of slices for XRD tests

\(\rho_w\): water density

\(A\) and \(B\): parameters for relation between \(\Theta\) and \(\chi\)

\(A'\): section area of the specimen

\(D\): soil water diffusivity \(=K(\Theta)\frac{d\theta}{dt}\)

\(d_{001}\): basal spacing of montmorillonite

\(G_s\): specific gravity

\(H=H(z, t)\): pressure head of the soil water

\(IA\): X-ray irradiated area by the incident wave

\(K(\Theta)\): hydraulic conductivity function of the soil

\(K_\text{V1}\): tested material Kunigel V1

\(L\): state of interlayer water molecule arrangement of montmorillonite. \(L=0w, 1w, 2w\) and \(3w: 0, 1, 2, 3\) rows of water molecules
$n$: a positive integer ($=1$)

$n'$: effective porosity ($=\Theta_s-\Theta$).

Phases I, II, III, IV: phases of swelling pressure development

$p_s$: swelling pressure

$Q$: amount of absorbed water

RH: relative humidity

$S_s$: degree of saturation

$t$: water absorption time

Types I and II: specimen installation types, see Fig. 4d and 3e

$v(z,t)$: volumetric flux or flow velocity of soil water

$w$: gravimetric water content

$w_{ini}$: $w$ under the air-dried condition

$z$: position, $z=0$ at bottom of the specimen and upwards as positive
1. Introduction

Geological disposal for the high level radioactive waste (HLW) is expected to be commercialized in many countries by the middle of the 21st century (METI 2018). In Japan, it has been decided that a disposal system will be placed in stable host rock formation more than 300 m underground (NUMO 2019). To guarantee the safety of the disposal system over a long timescale (i.e. several thousands to several tens of thousands years), a multi-barrier system consisting of engineered and natural barriers is under consideration as shown in Fig. 1 (JNC 1999, p IV-63). In such a system, bentonite is selected as a candidate material for buffer and backfill materials because of its excellent natural properties after being heavily compacted. One crucially important periods for this system is expected to be the saturation process during which ground water fills voids in the initially unsaturated buffer and backfill materials and engineering gaps of the disposal pits. This period is expected to last approximately 50-100 years, during which the surrounding temperature might reach a maximum level (e.g. a maximum temperature of 100 °C is a value for design in Japanese project) because of the extensive release of the radioactive energy from the waste (JNC 1999, p IV-94). Concerning on the saturation process of buffer and backfill materials, the water movement behaviors in the compacted bentonite is studied.

For most bentonite candidates for buffer materials, montmorillonite, a clay mineral belonging to smectite group, is the main component: it makes the bentonite swell when wetted. The swelling feature of montmorillonite results from penetration of water molecules into the 2:1 crystalline layer structure of montmorillonite. Consequently, at least three types of water exist in the compacted bentonite: interlayer water in montmorillonite, absorbed water sounding the outside surface of montmorillonite, and free water in voids between soil particles. Movements of interlayer water and water of all kinds (global water) are discussed in this study.

Movement of interlayer water has been extensively studied experimentally (Moore and Hower 1986; Watanabe and Sato 1988; Iwasaki and Watanabe 1988; Olis et al. 1990; Sato et al. 1992; Yamada et al. 1994; Fernandez et al. 2004; Morodome and Kawamura 2009) and numerically (Ferrage et al. 2005; Holmboe et al. 2012). However, in the past studies, water vapor with controlled relative humidity (up to 100%) rather than liquid water was mostly used to cure powder bentonite rather than compacted one. Under such testing conditions, maximum water content of cured bentonite was limited and the
interlayer movement might be different between powder and compacted bentonites. Additionally, in the above mentioned works, no attempt was made to correlate swelling pressure and interlayer water movement. In this study, a testing procedure is designed to measure the swelling pressure development when it is wetted, and then to measure the interlayer water movement of the bentonite. Discussions are made on the relation between development of swelling pressure and movement of interlayer water. For the movement of global water, some students were conducted to obtain the diffusivity of water in compacted bentonite (Takeuchi et al. 1995; Chijimatsu et al. 2000; Komine et al. 2018). In this work, diffusivity was also studied in the same manner, while a different calculation method is proposed, which provides good accuracy and simple calculation.

2. Testing program and methodology

2.1 Testing material

A commercial bentonite, Kunigel V1 (K_V1) produced from Tsukinuno mine, Japan, was used for this study. The swelling characteristics of K_V1 has been widely investigated (Komine and Ogata 1994; Sun et al. 2009 and among others). Its main mineral components were reported by Ito et al. (1993, 1994) based on several analytical methods. Their results were reproduced in Table 1. It is apparent that montmorillonite and chalcedony occupy more than 80% by mass. The cation exchange capacity (CEC) and amount of extractable cations (EXC) are also listed in Table 1, which were obtained base on method of JGS (2009) while considering the results by Okazaki et al. (1961) and Rollins and Pool (1968). It can be seen that Na\(^+\) and Ca\(^{2+}\) are dominant in K_V1. The specific gravity \((G_s)\) shown in Table 1 is an average value of six data obtained from two independent tests according to (JIS 2009a).

It is not straightforward to measure the gradation curve of K_V1 by the settlement method following JIS (2009b), which bases on the Stokes' law. JIS (2009b) recommends a mass of ~50 g for clayey material to mix with water for a 1 L solution. Tests by varying the mass of K_V1 from 5-36g were conducted and the results are plotted in Fig. 2. It can be seen that the passing percentage exceeds 100% significantly for 36g cases, which indicates that Stokes' law may be not applicable in these cases. Curves obtained by 15-30g cases seem reasonably consistent and repeatable, while curves deviate when further reducing mass to 5-10g. Two points obtained by Ito et al. (1994) with settlement and sieving methods are close to 20g cases. They pointed out about 5% variation affected by testing methods. An average line based on 15-30g cases is recommended herein.
Fig. 3 depicts an XRD profile of an air dried K_V1 specimen compacted to a dry density ($\rho_d$) of 1.45 Mg/m$^3$. The laboratory environments for air drying are controlled for a relative humidity RH of ~50% and temperature of ~23°C. The corresponding minerals of peaks in Fig. 3 are determined based on Komine and Ogata (1994), Komine (2004), and Ito and Komine (2008). The peak near $2\theta=7^\circ$ is the reflection of 001 surface of montmorillonite and is the object detailed observed in this study. It is noteworthy that the peaks indicated by quartz might also be chalcedony because their reflection positions are the same (Ito et al. 1994).

2.2 Test apparatus, procedure, and programs

Swelling pressure and XRD tests were conducted to measure interlayer and global movements of water and swelling pressure ($p_s$). The apparatus used to measure swelling pressure of compacted K_V1 powder is shown in Fig. 4. Fig. 4a shows a traditional swelling pressure apparatus by which the vertical and lateral displacements of the specimen can be constrained by clamps and a metal mold, respectively. In this study, the metal mold was changed to a multi-ring mold (Fig. 4b and 4c). The multi-ring mold was composed of five mold rings (thickness: 2 mm), and four two-piece rings (thickness: 0.25–0.30 mm) installed between mold rings. The specimen was prepared by statically compressing the air dried K_V1 powder to 28 mm in diameter and 11 mm in height directly in the multi-ring mold. Then the specimen was installed to the apparatus in the mode of either Fig. 4d or Fig. 4e. The two methods were tried to access any effect of mold wall friction on test results. For Fig. 4d, an extra mold was mounted on the multi-ring mold to seal the bottom side of multi-ring mold by O-ring. For Fig. 4e, a larger cap and a filter paper were mounted directly on multi-ring mold and a soft silicone O-ring was used at bottom for sealing, by which the side wall friction of the mold was expected to be alleviated. In both cases, an outer ring was used to avoid squeezing out of two-piece rings during the test. After installing the specimen, a small vertical stress was applied for contact. After fixing the clamp, $p_s$ and vertical displacement were recorded while supplying water from the bottom for a certain time period. The specimen conditions and water absorption time ($t$) are shown in Table 2. Nine specimens were prepared for $t$ varying from ~2 to ~450 hr. The average value of the air dried water content ($w_{\text{ini}}$) of the bentonite powder before compression was 7.9% and the average initial dry density after compression ($\rho_{\text{dini}}$) was 1.54 Mg/m$^3$. The specimen volume would generally swell slight during water absorption because of the system compliance, although final dry densities after water absorption ($\rho_{df}$) of two specimens ($t$ = 139 and 188 hr.) were found to increase according to vertical displacement measurements.
After water absorption, the multi-ring mold was taken out and gripped to constrain specimen deformation (Fig. 5a). Then, two-piece rings were removed and the specimen was sliced carefully one-by-one into five slices by using a thin saw (thickness: 0.20-0.25mm). The slices were then immediately sealed by Parafilm for at least 24 hr. under the constrained condition. It is noteworthy that even though much care was taken to reduce specimen disturbance and deformation, a certain amount of swelling or elastic rebound or both might occur during slicing work. The XRD tests were conducted on 45 slices under the instrumental setup of XRD equipment (RIGAKU RINT-Ultima III) shown in Fig. 5 and Table 3. As shown in Table 3, X-ray scan range was set as 2.7-20° with the scanning speed of 10°/min (i.e. <2 min for one slice). The testing duration was limited to the greatest extent possible because w may change and non-uniform deformation of slices might occur in the laboratory environment. The employment of a 1-D detector with 128 strips enhanced the scanning speed and guaranteed a sufficient reflected intensity. The irradiated area (IA) on the slices by the X-ray incident wave increases as diffraction angle 2θ (the angle between incident and reflected waves) decreases. An example of the calculated IA is shown in Fig. 5b-5e. IA slightly exceeds the area of the slice for 2θ=2.7°-3.43°, which would reduce the reflection intensity. Because 2θ of the peak interested is generally larger than 3.43°, it poses no issue for current study.

3. Testing results

3.1 Development of swelling pressure

The measured $p_s$ is shown in Fig. 6 and $t$ is shown in the legend. It can be seen from Fig. 6a that $p_s$ curves are generally converged to 0.8-0.9 MPa for specimens with $t>100$ hr., although the case for $t=453$ hr. deviates from others somewhat. Values of $p_s$ with $t$ up to 50 hr. are shown in Fig. 6b, from which it can be seen that $p_s$ after passing their maxima varies in a band of 0.7-0.8 MPa, except the case of $t=43.1$ hr. Additionally, it can also be seen that $p_s$ curves gather to two groups for $t<5$ hr. and the three specimens with gentle increasing slopes are those with the specimen installation type I (Fig. 4d and Table 2). This fact implies that for instillation type I, measurement may be delayed because of the side wall friction. However, the side wall friction seems not to affect the final magnitudes of $p_s$.

The variation of $p_s$ curves of similar specimens in Fig. 6 is partially because some detailed optimizations of test procedures conducted between tests. On the other hand, it is indeed a task to produce technically identical $p_s$ curves especially as $\rho_d$ increases (Pusch 1980;
Sridharan et al. 1986; Komine 2004; Tanai et al. 2010; Sun et al. 2013; Wang et al. 2017). The variation of $p_s$ may be induced by very slight differences between tests such as initial dry densities of specimens, compliance of the system, etc., since $p_s$ may significant change accompanying a slight deformation (Komine 2004; Tanaka 2011).

From Fig. 6a, it can be seen that $p_s$ development can be divided roughly into four phases: I, the $p_s$ initial increase phase from beginning of water absorption until ~18 hr.; II, the $p_s$ reduction phase until ~70 hr.; III, the $p_s$ re-increase phase until ~130 hr.; and IV, the $p_s$ steady state phase from ~130 hr. In the Discussion section, some attempt is made to connect this apparent phenomenon to the water molecule state in montmorillonite.

3.2 Water content distributions

The distributions of $w$ and degree of saturation ($S_r$) in specimen after water absorption are shown respectively in Fig. 7a and 7b, respectively. It can be seen that in general, $w$ and $S_r$ increase as the position close to the water supply end and as $t$ increases. It seems that $w$ and $S_r$ are in a steady state for specimens with $t>139$ hr. (note steady state herein stands for a state that no further increase trend of $w$ can be observed). However, the interesting points is that $S_r$ in many positions exceeds 100% significantly (i.e. up to ~130%). Note that $S_r$ was calculated based on $w$ measured by a balance with 0.1 mg accuracy, $G_s$ (=2.792-2.799) and dry density after water absorption ($\rho_{df}$), which do not scatter large enough to explain $S_r>100\%$ issue fully.

Some attribute $S_r>100\%$ issue to water density $\rho_w>1$ Mg/m$^3$. The interlayer water density in bentonite has been disputed for decades, which was said to be in a range from 0.7 to 1.4 Mg/m$^3$ (Pusch et al. 1990). For compacted bentonite, Jacinto et al. (2012) claimed that water density in Na-montmorillonite may reach 1.32 Mg/m$^3$ because $S_r$ values of their two compacted bentonite specimens were up to 117% if $\rho_w=1$ Mg/m$^3$, and Komine (2018) expressed the same idea since their $S_r$ values ranged in 105%-121% for three types of bentonites in 17 compacted specimens with $\rho_s$ of 1.21-1.85 Mg/m$^3$. The mechanism of intermolecular and surface forces also shown that water density may vary significantly from bulk water when it is sandwiched between two plates as the distance between plates becomes very close, i.e., Nano- or angstrom-level (Israelachvili 2011). Calculation of $S_r$ in Fig. 7b was based on $\rho_{df}$, while calculation based on dry density of XRD slices ($\rho_{XRD}$), which is the density after slicing and trimming, is shown in Fig. 8. It can be seen that $S_r \leq100\%$ with only one apparent exception. The difference of dry densities between $\rho_{df}$ and $\rho_{XRD}$ (negative sign for a small $\rho_{XRD}$) is plotted in Fig. 9, which demonstrates that in general dry density declines, especially at lower parts of specimens after slicing. Fig. 7
and Fig. 8 imply that interlayer water molecules with \( \rho_w > 1 \text{ Mg/m}^3 \) may change to a state or arrangement like bulk water with \( \rho_w = 1 \text{ Mg/m}^3 \) during swelling or elastic rebounding after their \( \rho_s \) was released. If it is true, if may be said that a saturated condition could not be reached from about 3 mm away from the water supply end even for \( t=453 \text{ hr.} \) (~19 days).

3.3 XRD diffractions for different water contents

Three typical results of XRD tests are shown in Fig. 10 (note the range of y-axis scales are all adjusted to the same; y-axis origins are shifted for clear visualization). It can be seen from Fig. 10 that the 001 peak of montmorillonite move gradually to small \( 2\theta \) as \( w \) increases and all the peaks move to \( 2\theta = -4.6^\circ \) for specimens of \( t=139 \text{ hr.} \). From the peak position, the basal spacing \( (d_{001}) \) of montmorillonite can be calculated by Bragg’s law:

\[
\text{Eqn. 1} \quad d_{001} = n\lambda/2\sin\theta_{\text{peak}}
\]

Therein, \( \lambda \): wavelength of incident wave (\( =1.5418 \text{ Å for Cu}\alpha \)), \( n \): a positive integer (\( =1 \)), \( \theta_{\text{peak}} \): \( \theta \) at peak maximum.

The relation between \( d_{001} \) and \( w \) is plotted in Fig. 11, in which data for oven (i.e. \( 110^\circ \text{C} \)) and air dried specimens and 10 other specimens cured under different relative humidity (RH) values are also added. It is readily apparent that \( d_{001} \) increases stepwise as \( w \) increases. This results are consistent with past studies (Moore and Hower 1986; Watanabe and Sato 1988; Iwasaki and Watanabe 1988; Olis et al. 1990; Sato et al. 1992; Yamada et al. 1994; Fernandez et al. 2004; Ferrage et al. 2005; Morodome and Kawamura 2009; Holmboe et al. 2012). Note that for the experimental works of past studies above, tested materials were not compacted generally and water vapor with controlled RH rather than liquid water was used to control water content. The general idea for the stepwise \( d_{001} \) is that each step represents a state of water molecule arrangement \( (L) \). As shown in Fig. 11 and Fig. 12, a step-up of \( d_{001} \) implies one more row of water molecule increase. Currently, it is generally agreed that there are four states in montmorillonite (i.e., \( L=0w, 1w, 2w \) and \( 3w \)) and \( d_{001} \) values shown in Fig. 12 are average values read from Fig. 11.

In past studies, \( d_{001} \) were mostly shown versus RH rather than \( w \). And the tested materials were carefully washed to purify the exchangeable ions (mostly purifying to Na-, Ca-, K- or Mg-montmorillonite). In this study, such a washing process was not conducted considering the practical purpose of bentonite for geological disposal. For comparison purposes, data of past studies for materials produced from the same mine as K_V1 (i.e. Tsukinuno mine, Japan) are summarized in Fig. 13 together with some data in this study.
Temperature was 50°C in Morodome and Kawamura (2009) and room temperature for others. It can be seen that $d_{001}$ of Ca-montmorillonite is one or two steps higher than that of Na-montmorillonite from RH=0% until 60-70%, and they converge thereafter. This observation implies that water molecules might be first absorbed into the interlayer space occupied by Ca$^{2+}$ ions mostly at low $w$, whereas once $w$ exceeds $w$ corresponding to RH=60-70%, the effect of ion types on $d_{001}$ might be minor. Data in this study generally follow steps of Na-montmorillonite except that at RH=~10%.

4. Discussion

4.1 Relation between $L$ states and $p_s$ phases

To date, the only information used from XRD tests is $\theta_{\text{peak}}$, which was often done in the past, because quantitative analyses for clay minerals are generally very complicated and difficult (Moore and Reynolds 1997; Drits and Tchoubar 1990; Dinnebier and Billinge 2008). In this section, the discussion is still limited to qualitative analysis, while efforts are made to extract a bit more information. It can be seen from Fig. 10 that values of $\theta_{\text{peak}}$ at 001 peaks of montmorillonite might be very close, however, the peak shapes (e.g. magnitude, width, symmetry, etc.) might be very different. The reason of shape difference is expected to be co-existence of $L$ states for a certain $w$ (Moore and Hower 1986; Ferrage et al. 2005; Morodome and Kawamura 2009; Holmboe et al. 2012). In other words, $d_{001}$ of montmorillonite might be not a single value at certain $w$: it changes depending on $w$ resulting in different peak shapes.

It was assumed that two $L$ states co-existed for a certain $w$, and the peak decomposition (i.e. profile fitting) was conducted to separate peaks corresponding to each $L$ states in a way illustrated by Fig. 14. First, the peak background (dashed line in Fig. 14a) was assumed as a linear line from the left peak tail (the bottom point of the curve valley) to the right peak tail. The background was subtracted from the profile, after which the resultant peak data were normalized by its intensity maximum, as indicated by hollow squares in Fig. 14b. For the normalized data, two peak positions near the $L$ state lines were designated arbitrarily as the initial condition for peak fitting. The normalized data was fitted using the composed line of two peaks by the least square method. Then the two peaks were regarded respectively as the corresponding peaks of their nearest $L$ states. Pearson VII function was used for the shape of two peaks because it is one of the most popular functions for XRD fitting (Wertheim et al. 1974; Hall Jr 1977; Langford 1978).
instrumental broadening (Klug and Alexander 1974; Bobert and Coelho 1992; Dinnebier and Billinge 2008; Ida et al. 2018) are not considered. And hence, the resultant peak positions might deviate from their corresponding $L$ state lines somewhat.

Results obtained after the above processes are portrayed in Fig. 15. It can be seen that $\theta_{\text{peak}}$ of decomposed peaks move to small $2\theta$ as $t$ increases until $t$ exceeds about 139 hr. (Fig. 15f). All $\theta_{\text{peak}}$ of decomposed peaks reach or exceed $L=2w$ state after $t=17.1$ hr., and $L=3w$ state after $t=139$ hr. Decomposed peaks seem to be in a stable state for specimens with $t>139$ hr. (i.e. no further movement trend towards small $2\theta$).

The corresponding $d_{001}$ of $\theta_{\text{peak}}$ of decomposed peaks are calculated and plotted together with development of $p_s$ as shown Fig. 16. It can be seen that phases of $p_s$ generally coincide with $L$ states, i.e., $p_s$ reaches its first peak (i.e. phase I) when $L$ moves from 1w to at least 2w throughout the specimen; $p_s$ decreases and re-increases (i.e. phases II and III) when $L$ moves from 2w to 3w; and $p_s$ reaches a steady state (i.e. phase IV) when $L=3w$. These observations imply that development of $p_s$ might be an apparent result of water molecule movement in the interlayer spaces. Currently, there is no data available to know if there are peaks at $2\theta$ smaller than 2.7° (=the minimum $2\theta$ in this study), and it cannot be said that the peaks measured in this study are the peaks before release of $p_s$ during water absorption since specimens apparently deformed during slicing work. However, from Fig. 16, it might be said that water penetration up to $L=3w$ should at least a partial reason of $p_s$ development. This is important because with a $d_{001}$ $\leq$20 Å, net force between crystalline layers would be attractive force base on diffuse double layer theory. In other words, $p_s$ measured in this ways might at least partially attribute to other mechanics, such as attractive and repel force oscillation when two planes are very close (Israelachvili 2011). On the other hand, there is not experimental evidence neither available to explain phases II (reduction phase) and III (re-increase phase) of $p_s$. It is expected that the reduction of $p_s$ might be related to collapse behavior of unsaturated soils when wetted. If so, measured $p_s$ would be a net swelling pressure resulted by swelling behavior of montmorillonite and collapse behavior of unsaturated soils.

4.2 Diffusivity for global water movement

With the water content distribution shown in Fig. 7, the global movement of water in the specimens may be evaluated in terms of diffusivity (Philip 1958; Takeuchi et al. 1995; Chijimatsu et al. 2000). The flow equation for the relation of volumetric water content ($\Theta$) – soil water diffusivity ($D$) is obtainable by combining the extended Darcy equation for unsaturated soil and the conservation of mass (Richards 1931; Klute 1972) as shown
in Eqns. 2-4. Then, Eqn. 4 changes to Eqn. 5 if the elevation potential part ($\partial K / \partial z$) can be ignored.

Eqn. 2 \( \mathbf{v}(z,t) = -K(\Theta)\left(\frac{\partial H}{\partial z} + 1\right) \)

Eqn. 3 \( \frac{\partial \Theta}{\partial t} = -\frac{\partial \mathbf{v}}{\partial z} \)

Eqn. 4 \( \frac{\partial \Theta}{\partial t} = \frac{\partial}{\partial z}\left(D\frac{\partial \Theta}{\partial z}\right) + \frac{\partial K}{\partial z} \)

Eqn. 5 \( \frac{\partial \Theta}{\partial t} = \frac{\partial}{\partial z}\left(D\frac{\partial \Theta}{\partial z}\right) \)

where \( \mathbf{v}(z,t) \): volumetric flux or flow velocity of soil water

\( H=H(z, t) \): pressure head of the soil water

\( K(\Theta) \): hydraulic conductivity function of the soil

\( z \): position, herein set \( z=0 \) at bottom of the specimen and upwards as positive

\( D \): soil water diffusivity = \( K(\Theta) \frac{d\Theta}{dH} \)

Eqn. 5 is of the same form as the nonlinear diffusion equation encountered in diffusion theory. Bruce and Klute (1956) and Jackson (1964) used the Boltzmann transform, of which \( D \) was set as a function of \( \chi = z/\sqrt{t} \). They obtained:

Eqn. 6 \( D(\Theta_z) = \frac{1}{2}\left(\frac{d\chi}{d\Theta}\right)_{\Theta_z} \int_{\Theta_i}^{\Theta_z} \chi d\Theta \)

with initial condition: \( \Theta=\Theta_i \) when \( z>0 \) and \( t=0 \), and boundary condition: \( \Theta=\Theta_s \) when \( z=0 \) and \( t\leq0 \). The subscripts \( i, s \) and \( z \) respectively denote initial (i.e. air dried) and saturated conditions and position. Also, \( (d\chi/d\Theta)_{\Theta_z} \) is the slope of relation between \( \chi \) and \( \Theta \) at \( \Theta_z \).

The relation between \( \Theta \) and \( \chi \) is plotted in Fig. 17a based on measurement in Fig. 7. It can be seen that the relation is of a semi-logarithmic form. Eqn. 7 is used for fitting the \( \Theta \) and \( \chi \) relation and \( D(\Theta_z) \) is expressed in Eqn. 8 based on Eqn. 6 and Eqn. 7.

Eqn. 7 \( \chi = A - B\ln(\Theta - \Theta_i) \)
Eqn. 8 \[ D(\theta_z) = \frac{AB}{2} - \frac{B^2}{2} (\ln(\theta_z - \theta_1) - 1) \]

It is worth mentioning that \( \theta_z \) in Eqn. 8 may larger than \( \theta_1 \) because, as discussed in Fig. 7, \( S_r \) of some data apparently exceed 100%. Takeuchi et al. (1995) also measured the \( w \) distribution of compacted K_V1 specimen with diameter of 20mm, height of 20mm and \( \rho_d \) of 1.6 Mg/m\(^3\) at \( t=4, 24, 48 \) and 72 hr. Eqn. 7 is also applied to their experimental data as shown in Fig. 17b. It can be seen that the data generally follow Eqn. 7, though some data near \( \theta=20\% \) deviate from the fitting line somehow.

The estimated \( D \) of K_V1 from Eqn. 8 is compared with results of past studies by Takeuchi et al. (1995) and Komine et al. (2018). Takeuchi et al. (1995) used Eqn. 9 to estimate \( D \), of which the change of \( \theta \) between two measurements was taken.

Eqn. 9 \[ v = D \frac{\partial \theta}{\partial z}, \quad D = \frac{1}{t_2 - t_1} \int_{z_1}^{t_2} \left( \theta(t) - \theta_{t_1} \right) dz \]

Komine et al. (2018) measured the relation between \( t \) and amount of absorbed water \( (Q) \) and derived Eqn. 10 claiming that there was a liner relation between \( \sqrt{t} \) and \( Q \) from the beginning of water absorption until somewhere close to saturation.

Eqn. 10 \[ Q = 2A' n' e^{\frac{D}{\sqrt{\pi t}}} \]

where, \( A' \): sectional area of the specimen, \( n' \): effective porosity (\( = \theta_s - \theta_i \)). Consequently, Eqn. 10 might be regarded as an average \( D \).

The relation between \( S_r \) and \( D \) is plotted in Fig. 18 because this relation is more readily available from Takeuchi et al. (1995). In Fig. 18, estimated \( D \) from Eqn. 8 for data by Takeuchi et al. (1995) are also added. It can be seen that calculated \( D \) by Takeuchi et al. (1995) was markedly scatter, and an approximate U-shape curve was drawn as a representative. On the other hand, calculated \( D \) by Eqn. 8 seems to monotonically decrease at upper part of the U-shape curve. Nevertheless, it seems that, in general, all estimated \( D \) values are similar in terms of magnitude.

Predictions for experimental data obtained by Takeuchi et al. (1995) and Komine et al. (2018) using \( D \) obtained from Eqn. 8 are shown in Fig. 19. Fig. 19a shows that the prediction matches experiments of specimen with \( t=4h \) best, whereas partial deviation from experimental data of the other three specimens is observed. The positions of
deviation match deviation positions shown Fig. 17b as expected. The measured $Q$ in Komine et al. (2018) was converted to average $S_r$, by which the final average $S_r$ was about 105%. Consequently, $S_r$ at different $z$ was predicted by setting a maximum $S_r$ of 105%. The result is shown in Fig. 19b, which indicates that the prediction well matches experimental data for the first about 50 hr. and that $S_r$ is underestimated by ~7% thereafter. All in all, it might be said that estimation method of $D$ (i.e. Eqn. 8) is easy and the predictions are reasonably good.

5. Conclusion

A testing procedure was proposed to study the movements of interlayer and global water movements in compacted bentonite as well as development of swelling pressure ($p_s$) when compacted bentonite specimens were wetted with water. In this procedure, bentonite powder was compressed in a newly introduced multi-ring mold. Then $p_s$ was measured during a certain water absorption time ($t$). Finally, the specimens were sliced carefully for X-ray diffraction (XRD) tests to elucidate the movement of water in the interlayer space of montmorillonite. Gravimetric water content ($w$) for all slices were also carefully measured to observe the evolution of $w$ distribution in the compacted bentonite as $t$ increased, based on which the global water movement in terms of diffusivity ($D$) was estimated.

From $p_s$ measurements, results demonstrated that $p_s$ development can be divided into four phases: I, $p_s$ initial increase phase from beginning of water absorption until ~18 hr.; II, $p_s$ reduction phase until ~70 hr.; III, $p_s$ re-increase phase until ~130 hr.; and IV, $p_s$ steady state phase from ~130 hr. From XRD tests, it was confirmed that there were four states of interlayer water molecule arrangement of montmorillonite ($L$): $L=0w, 1w, 2w$ and $3w$, which respectively stand for 0, 1, 2, 3 rows of water molecules in the interlayer space of montmorillonite. Qualitative analysis on 001 peaks of montmorillonite obtained by XRD tests was conducted to decompose $L$ states under a certain $w$ condition. Results shown an apparent relation between phases of $p_s$ development and evolution of $L$ states: $p_s$ reached its first peak in phase I when $L$ moved from 1w to at least 2w throughout the specimen; $p_s$ decreased and re-increased in phases II and III when $L$ moved from 2w to 3w; and $p_s$ reached a steady state in phase IV when $L=3w$. This result implied that $p_s$ would be a net swelling pressure resulted by swelling behavior of montmorillonite and collapse behavior of unsaturated soils, and mechanisms other than diffuse double layer theory might also contribute to $p_s$ measured.

Measurements of the $w$ distribution revealed that the degree of saturation ($S_r$) of
compacted bentonite exceeded 100% by a considerable degree. This result may be due to
that water density ($\rho_w$) in the compacted bentonite might be larger than 1 Mg/m$^3$. Based
on the $w$ distribution data, $D$ of compacted bentonite was estimated following the method
employing the Boltzmann transform. Comparisons between this and past studies implied
that $D$ calculated using the method adopted for this study well matched data from past
studies, and the method is rather easy to handle.

Acknowledgements

A part of the present work was performed as a part of activities of Research Institute of
Sustainable Future Society, Waseda Research Institute for Science and Engineering,
Waseda University. Part of this study was supported by the Ministry of Economy, Trade
and Industry (METI) of Japan. The authors would like to thank Dr. Y. Watanabe and
Dr. S. Yokoyama of the Central Research Institute of Electric Power Industry, Japan for
their valuable comments related to swelling pressure and XRD tests. All the XRD tests
were conducted at the Materials Characterization Central Laboratory, Waseda University
(Izutani et al. 2016).
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Table 1 Main mineral components and physical properties of K_V1

| Main mineral components, their standard formula and contents in weight from Ito et al. (1993 and 1994) | Montmorillonite | Quartz | Chalcedony | Plagioclase | Calcite | Dolomite | Analcime | Pyrite |
|---|---|---|---|---|---|---|---|---|
| Montmorillonite | SiO₂ | - | - | - | - | - | - | - |
| Quartz | SiO₂ | 0.5-0.7% | 0.5-0.7% | 0.5-0.7% | 0.5-0.7% | 0.5-0.7% | 0.5-0.7% | 0.5-0.7% |
| Chalcedony | SiO₂ | 37-38% | 37-38% | 37-38% | 37-38% | 37-38% | 37-38% | 37-38% |
| Plagioclase | Na₂O·Al₂O₃·6SiO₂ | 2.7-5.5% | 2.7-5.5% | 2.7-5.5% | 2.7-5.5% | 2.7-5.5% | 2.7-5.5% | 2.7-5.5% |
| Calcite | CaCO₃ | 2.1-2.6% | 2.1-2.6% | 2.1-2.6% | 2.1-2.6% | 2.1-2.6% | 2.1-2.6% | 2.1-2.6% |
| Dolomite | CaMg(CO₃)₂ | 2.0-2.8% | 2.0-2.8% | 2.0-2.8% | 2.0-2.8% | 2.0-2.8% | 2.0-2.8% | 2.0-2.8% |
| Analcime | NaAlSi₂O₆·H₂O | 3.0-3.5% | 3.0-3.5% | 3.0-3.5% | 3.0-3.5% | 3.0-3.5% | 3.0-3.5% | 3.0-3.5% |
| Pyrite | FeS₂ | 0.5-0.7% | 0.5-0.7% | 0.5-0.7% | 0.5-0.7% | 0.5-0.7% | 0.5-0.7% | 0.5-0.7% |
| Total cation exchange capacity (CEC) and amount of extractable cations (EXC)* (cmol/kg) *EXC includes exchangeable and water-soluble cations | CEC | 66.8 | 66.8 | 66.8 | 66.8 | 66.8 | 66.8 | 66.8 |
| | EXC (Na⁺) | 57.1 | 57.1 | 57.1 | 57.1 | 57.1 | 57.1 | 57.1 |
| | EXC (Ca²⁺) | 35.5 | 35.5 | 35.5 | 35.5 | 35.5 | 35.5 | 35.5 |
| | EXC (K⁺) | 1.8 | 1.8 | 1.8 | 1.8 | 1.8 | 1.8 | 1.8 |
| | EXC (Mg²⁺) | 1.3 | 1.3 | 1.3 | 1.3 | 1.3 | 1.3 | 1.3 |
| Specific gravity Gₛ | 2.795 (min:2.792-max:2.799) | 2.795 (min:2.792-max:2.799) | 2.795 (min:2.792-max:2.799) | 2.795 (min:2.792-max:2.799) | 2.795 (min:2.792-max:2.799) | 2.795 (min:2.792-max:2.799) | 2.795 (min:2.792-max:2.799) | 2.795 (min:2.792-max:2.799) |
Table 2 Specimen conditions for the swelling pressure tests

| Water absorption time $t$ (hours) | Air dried water content $w_{\text{ini}}$ (%) | Initial dry density $\rho_{\text{ini}}$ Mg/m$^3$ | Final average water content $w_f$ (%) | Final dry density $\rho_d$ Mg/m$^3$ | Specimen Installation type |
|----------------------------------|---------------------------------------------|-----------------------------------------------|---------------------------------------|------------------------------------|--------------------------|
| 1.96                             | 8.11                                        | 1.55                                          | 13.31                                 | 1.54                               | II                       |
| 4.37                             | 8.33                                        | 1.53                                          | 15.32                                 | 1.51                               | II                       |
| 17.1                             | 8.24                                        | 1.56                                          | 19.26                                 | 1.55                               | I                        |
| 43.1                             | 9.65                                        | 1.54                                          | 23.38                                 | 1.52                               | I                        |
| 65.7                             | 7.98                                        | 1.53                                          | 27.37                                 | 1.52                               | II                       |
| 139                              | 7.91                                        | 1.54                                          | 29.61                                 | 1.56                               | II                       |
| 188                              | 8.09                                        | 1.53                                          | 30.55                                 | 1.54                               | I                        |
| 327                              | 7.88                                        | 1.53                                          | 29.47                                 | 1.51                               | II                       |
| 453                              | 8.07                                        | 1.56                                          | 30.59                                 | 1.54                               | II                       |
| Average                          | 8.25                                        | 1.54                                          | -                                     | 1.53                               | -                        |
| Min.                             | 7.88                                        | 1.53                                          | -                                     | 1.51                               | -                        |
| Max.                             | 9.65                                        | 1.56                                          | -                                     | 1.56                               | -                        |
| **Table 3** Instrumental setup for XRD test |
|-------------------------------------------|
| **Goniometer** | Bragg-Brentano geometry (radius: 285mm) |
| **R-ray source** | Ni-filtered CuKα radiation (40kV 40mA) |
| **Incident beam side** | Soller slits | ±2.5° |
| | Divergence slit (DS) | 0.125° |
| **Detector side** | Soller Slits | ±2.5° |
| | Scattering slits (SS) | Open |
| | Receiving slit (RS) | Open |
| **Scanning conditions** | type | Continuous |
| | speed | 10°/min |
| | step | 0.02° |
| | range | 2.7-20° |
| **Detector** | 1-D detector |
| | 128 strips with width of 0.1mm |
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Fig. 11 Relation between \( w \) and \( d_{001} \)

![Graph showing the relation between water content \( w \) and basal spacing \( d_{001} \).](image)

- \( L=0w \) with \( d_{001} \approx 10.1 \AA \)
- \( L=1w \) with \( d_{001} \approx 12.5 \AA \)
- \( L=2w \) with \( d_{001} \approx 15.5 \AA \)
- \( L=3w \) with \( d_{001} \approx 19.0 \AA \)

Fig. 12 Schematic illustration of the state of water molecule arrangement (\( L \))

![Schematic illustration showing different states of water molecule arrangement.](image)

- Water
- 2:1 sheet structure of montmorillonite
- Exchangeable cations

\( w = 8.51\% \) for air dried specimen
\( w = 6.58\% \) for RH = 58.4\%
Fig. 13 Comparison of RH and $d_{001}$ relation between purified montmorillonite and K_V1 used for this study.
Fig. 14 Data process for XRD measurement

(a) Raw data and background

(b) Normalized data and decomposition

XRD intensity ($\times 10^4$ CPS)

Diffraction angle $2\theta$ (°)

Assumed background

Peak reflected by 001 surface of montmorillonite

Composition of two peaks

L=3w  L=2w  L=1w  L=0w
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