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ORIGINAL PAPER

Predicting Swelling Behavior of a Na^+-Bentonite Used in GCLs

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Abstract

The swelling behavior of a Na^+-bentonite used in geosynthetic clay liners with lower dry unit weights (less than about 11 kN/m^3) was investigated experimentally, and analyzed by diffuse double layer (DDL) theory. Four types of liquid were used. It has been found that the measured swelling pressures are about 20% of that predicted by DDL theory. There are linear relationships between the calculated double layer thickness and the measured corresponding free swelling index and liquid limit.

Keywords Bentonite · Geosynthetic clay liner · Swelling pressure · Diffuse double layer

Introduction

Geosynthetic clay liner (GCL) has been widely used in constructing liner systems of waste disposal landfills and water storage pools because of its lower permeability as well as self-healing capacity [1–4]. While the permeability of a GCL as well as self-healing capacity depends on the swelling behavior of the bentonite used in it [5, 6]. Therefore, if the swelling behavior of bentonites used in GCLs can be predicted, it will be very useful for design GCL liners.

In predicting the swelling pressure of bentonites, the basic theory used is the diffuse double layer (DDL) theory which is also known as the Gouy-Chapman theory [7, 8]. The explicit equations of DDL theory were derived assuming parallel alignment of two platelet soil particles, but in a mass of bentonite there may be both parallel alignments of platelets, and also face-to-edge configurations [9]. In the literature, there are reported results demonstrating that DDL theory, with certain modifications, can be used to predict the swelling pressure of bentonite [10, 11], but there are also reported results showing that there are considerable differences between the theoretical predictions and experimental results [12].

Most of the research results available in the literature concerning swelling behavior of bentonite are focused on its application in nuclear waste disposal [13], in which the bentonite has a relatively high compacted dry unit weight. For the bentonite used in GCL, normally it is in a relatively loose state with a dry unit weight about 10 kN/m^3. There are very limited researches quantitatively evaluating the applicability of DDL theory to bentonites with relative lower density.

In this study, the swelling behavior of a bentonite used in a GCL was investigated experimentally using four different liquids. The swelling behavior of the bentonite was predicted using DDL theory, and compared with the measured data. The applicability of DDL theory to bentonites at relatively low densities, such as used in GCLs, is then discussed.

Experimental Investigation

Materials Used and Test Methods

Based on the information from the manufacturer of the GCL, the bentonites were mined at two locations (Colony and Lovell) in Wyoming, USA, and their average main chemical compositions and mineral contents are given in Table 1.

Considering the cations existed in the leakage of landfill as well as easy to handle in the laboratory, the four liquids selected for this study are: (1) a tap water (Saga City, Japan); (2) 0.17 mol/l NaCl solution; (3) 2.17 mol/l ethanol solution;

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Table 1 Chemical compositions and minerals of the bentonites (X-ray semi-quantitative analysis) (The data are provided by the manufacturer)

Sampling location		Content[a]
Chemical composition (% by weight)	Si as SiO_2	65.19
	Al as Al_2O_3	20.86
	Ca as CaO	0.94
	Na as Na_2O	2.31
	Mg as MgO	2.43
	Fe as Fe_2O_3	1.80
	K as K_2O	0.30
	Cr as Cr_2O_3	0.02
	Mn as MnO	0.06
	Ti as TiO_2	0.15
	V as V_2O_5	0.04
Minerals (% by weight)	Quartz	3
	Potassium feldspar	Trace
	Plagioclase feldspar	2
	Calcite	Trace
	Opal	4
	Clinoptilolite	Trace
	Dioctahedral smectite	88
	Illite	3

[a]Average values of the bentonites from Colony and Lovell in Wyoming, USA

and (4) 0.1 mol/l $CaCl_2$ solution. For each liquid, the liquid limit (w_l), plastic limit (w_p), and free swelling index (FSI) of the bentonite, and the pH of the liquid phase from the soil–water mixture at a solid/liquid ratio of 1:10 were measured. The results are summarized in Table 2. Values of FSI were determined according to ASTM D 5890-11 [14].

The swelling pressures (p_m) were measured using a modified odometer device under a constant volume condition, which is similar to Method-C of ASTM D 4546-96 [15]. The test procedure is as follows.

(1) Put the bentonite with an initial water content of about 10–30% into a mold of 60 mm in diameter and 20 mm in height. Apply a vertical pressure of 50 to 1000 kPa for 2 h to compress the sample. By this way, soil samples with different dry unit weight were produced. The resulting samples (after release of the compression pressure) had dry unit weights of 7.45–10.78 kN/m^3.
(2) Cut the sample into 5 mm in thickness and reset into the equipment for swelling pressure test.
(3) Set a load cell between the soil sample and the reaction frame for measuring swelling pressure, and a dial gauge for monitoring vertical displacement of the sample.
(4) Put desired liquid and start the test. During swelling pressure test, the thickness of the soil sample was not allowed to change. Possible vertical displacement of the sample due to the deformation of load cell was eliminated by manually adjustment.
(5) When the swelling pressure stabilized, the test was terminated and the final water content of the soil sample and the mass of the sample were measured. In the analysis, the final water content was used.

Table 2 Liquid limit (w_l), plastic limit (w_p) and free swelling index (FSI)

Fluid type	pH	Liquid limit (w_l), %	Plastic limit (w_p), %	FSI ml/2 g
Tap water	7.02	537	45.8	30.0
0.17 mol/l NaCl solution	7.24	235	46.3	16.5
2.17 mol/l ethanol solution	7.46	560	67.4	30.0
0.1 mol/l $CaCl_2$ solution	7.60	165		9.0

Typical Swelling Pressure-Elapsed Time Curves

Four swelling pressure tests were conducted using the tap water, three tests for the 0.17 mol/l NaCl solution and three tests for the 2.17 mol/l ethanol solution. The results of using the tap water are depicted in Fig. 1. The measured final water content—swelling pressure relationships are shown in Figs. 2, 3 and 4 for using the tap water, 0.17 mol/l solution and 2.17 mol/l ethanol solution respectively. As for using 0.1 mol/l $CaCl_2$ solution, with an initial dry unit weight of about 10 kN/m^3, the bentonite almost not swelled (measured swelling pressure was close to zero).

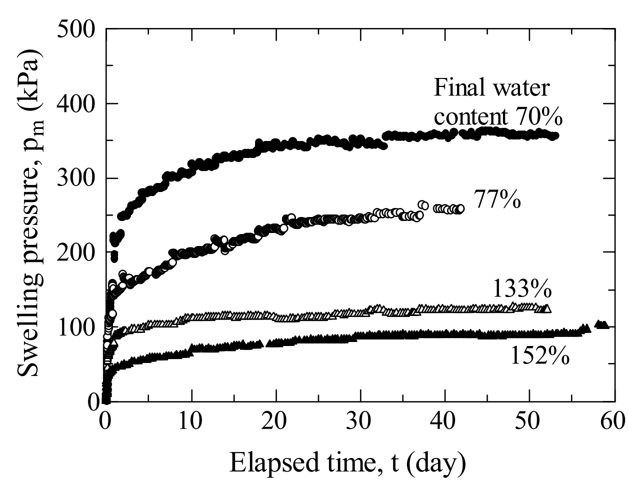


Fig. 1 Swelling pressure versus elapsed time when using tap water

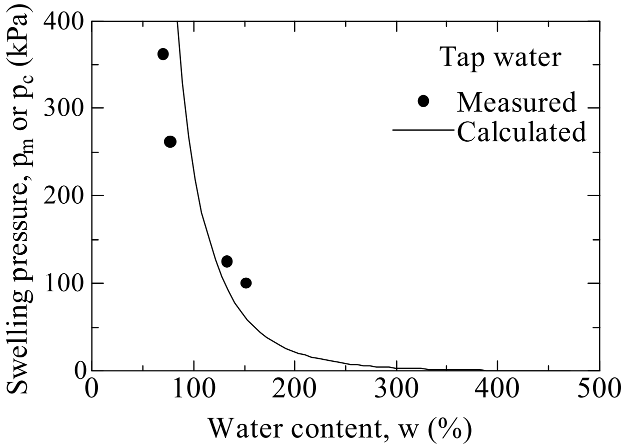


Fig. 2 Comparison of swelling pressures using tap water

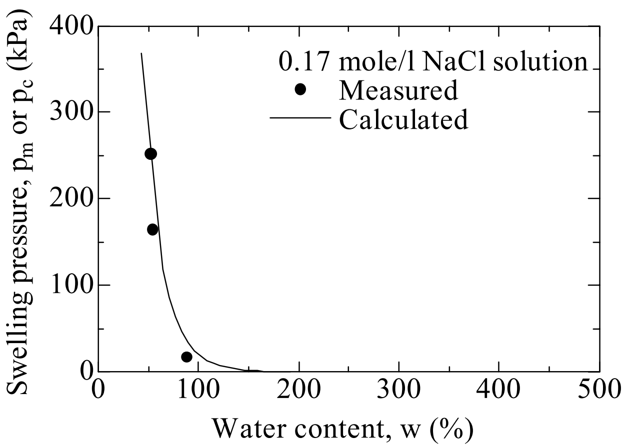


Fig. 3 Comparison of swelling pressures using 1% NaCl

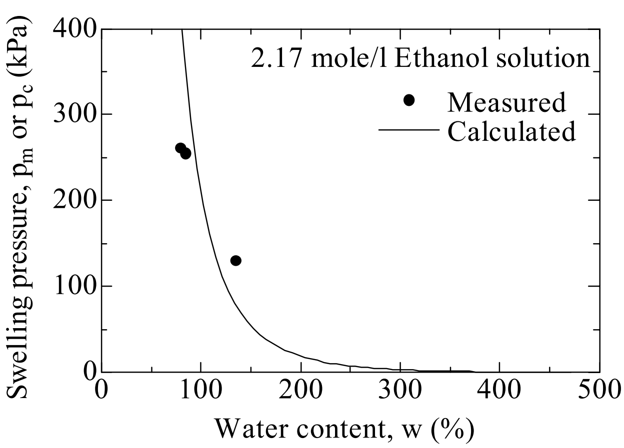


Fig. 4 Comparison of swelling pressures using 10% ethanol solution

Repulsion and Attraction Forces Between Clay Particles

DDL Theory

Basic Equations

From the DDL theory [7, 8], repulsive force (p) per unit area between two parallel clay platelets can be calculated as follows:

$$p_R = 2nkT[\cosh u - 1] \tag{1}$$

where n = molar concentration of cations in pore fluid (mole/m^3 multiplied by Avogadro's number $N_A = 6.023 \times 10^{23}$), k = Boltzmann's constant ($= 1.38 \times 10^{-23}$ J/K), T = absolute temperature in Kelvins, and u = electrical potential at the centre of two parallel clay platelets and can be calculated as:

$$u = 8\tanh^{-1}\left[\exp(-\kappa \cdot d) \cdot \tanh(z/4)\right] \tag{2}$$

where d = half the distance between two parallel clay platelets, κ = double layer parameter ($1/\kappa$ = thickness of the double layer), and z = potential at the surface of a clay particle. κ and z can be calculated as follows:

$$\kappa = \sqrt{\frac{2(e')^2 \nu^2 n}{\varepsilon kT}} \tag{3}$$

$$z = 2\sinh^{-1}\left(96.5 \times \frac{\mathrm{CEC}}{S}\sqrt{\frac{1}{8\varepsilon nkT}}\right) \tag{4}$$

where ν = valence of cation, e' = elementary electric charge ($= 1.602 \times 10^{-19}$ C), ε = dielectric constant of pore fluid (for water, $\varepsilon = 7.083 \times 10^{-10}$ C^2/J/M, and for ethanol, $\varepsilon = 2.15 \times 10^{-10}$ C^2/J/M), CEC = cation exchange capacity of the clay (meq/u$_a$, where u$_a$ is atomic mass unit, 1u$_a$ = 1.6605×10^{-24} g, and 1 meq = 1.602×10^{-22} C), and S = specific surface area of the clay (m^2/g). In Eq. (4), 96.5 is a multiple for converting meq/u$_a$ to C/g.

Method for Considering Multiple Cations

The equations given above are for the case of a single cation. However, for most real situations, there are multiple cations in the pore liquid. Sridharan and Jayadeva [16] proposed that: the value of n can be summed up for cations existed in the liquid; and the value of ν can be calculated as the weighted average value using the value of molar concentration. This method is adopted in this study.

Estimating Dry Unit Weight and Water Content

It is assumed that the volume of voids in a soil sample can be estimated as the specific surface area multiplied by half the distance between adjacent parallel clay platelets (d) [16]. Then the relationship between dry unit weight (γ_d) and d is:

$$\gamma_{\mathrm{d}} = \frac{G \cdot \gamma_w}{1 + G \cdot \gamma_w S \cdot d} \tag{5}$$

and the water content is:

$$w = d \cdot S \cdot \gamma_{\mathrm{w}} \tag{6}$$

where G = specific gravity of clay (bentonite) particle, and γ_w = unit weight of water.

van der Waals Force

Considering the effect of retardation within soil particles, the van der Waals force between two parallel plates is calculated as [17]:

$$p_{\mathrm{A}} = \frac{A}{24\pi}\left(\frac{1}{d^3} + \frac{1}{(d+t)^3} - \frac{2}{(d+t/2)^3}\right) \tag{7}$$

where A = Hamaker constant. For montmorillonite (main mineral in the bentonite), A = 2.2×10^{-20} J [18] and t = thickness of the plates.

Net Repulsion Force

The net repulsion force (p_{N}) between two parallel clay particles can be calculated as:

$$p_{\mathrm{N}} = p_{\mathrm{R}} - p_{\mathrm{A}} \tag{8}$$

Measured and Predicted Results

Double Layer Thickness

The quality of the tap water in Saga City, Japan was checked by Saga City Waterworks and Sewerage Bureau (SCWSB) and published monthly. Based on the information published by SCWSB [19], the ranges and average concentrations of some main ions in and values of pH of the tap water of Saga City in 2015 are listed in Table 3. The sums Ca^{++} and Mg^{++} were published but most of them were Ca^{++} (personal communication with a staff in SCWSB). For simplicity assuming all of it is Ca^{++} (molar weight of 40 g), the molar concentration will be about 0.0006 mol/l. Further, Na^+ in the liquid phase of the bentonite and the tap water mixture with a solid/liquid ratio of 1:10, was measured as 590 mg/l. Referring to these values, it is assumed that Na^+ at a concentration of 590 mg/l (0.026 mol/l) and Ca^{++} of 26.8 mg/l (0.0006 mol/l) were present in all four types of liquid tested. The dielectric constant for the tap water, the 0.17 mol/l NaCl solution and the 0.1 mol/l $CaCl_2$ solution is the same as for pure water, and for the 2.17 mol/l ethanol solution a weighted average value of pure water and ethanol is used. The values of total molar concentrations of cations, weighted average valances and dielectric constants are listed in Table 4. The value of *CEC* measured using atomic absorption spectrophotometry is 77 cmol/kg (77 meq/100 g). The value of the specific surface area (S) of the bentonite has not been directly measured. The bentonite mined in Wyoming mainly consisted dioctahedral smectite mineral, and its S value is about 750 m^2/g [20]. Further based on the data in Table 1, the dioctahedral smectite content in the bentonite was about 88% of the total weights. Assuming an average dioctahedral smectite content of 85%, an S value of 637 m^2/gr. can be estimated, which is the value used in this study.

Using Eq. (3), the calculated values of $1/\kappa$ are listed in Table 4. Although the value of $1/\kappa$ cannot be directly measured, it is considered that the value of $1/\kappa$ should be related to the values of the FSI or w_l of the bentonite. The relationships between $1/\kappa$ and the measured FSI and w_l are plotted in Figs. 5 and 6 respectively. Generally, almost linear

Table 4 Properties of the fluids and calculated thickness of DDL

Parameters	Tap water	1% NaCl solution	10% Ethanol solution	1.1% $CaCl_2$ solution
n (mol/l)	0.0266[a]	0.1976	0.0266	0.1266
ν	1.023[b]	1.003[b]	1.023[b]	1.795[b]
ε ($\times 10^{-10}$ C^2/J/M)	7.083	7.083	6.590	7.083
$1/\kappa$ (Å)	18.2	6.8	17.6	4.8
Water content of water in DDL (%)	115.9	44.0	112.1	30.6

[a] 590 mg/l of Na^+ in the bentonite and the tap water mixture (solid/liquid ratio: 1:10) and 26.8 mg/l Ca^{++} in the tap water

[b] Weighted average of Na^+ and Ca^{2+}

Table 3 Ion concentrations in the tap water of Saga City, Japan

	Na^+ (mg/l)	Cl^- (mg/l)	Ca^{++} and Mg^{++} (mg/l)	pH	Remark
Average	7.6	10.4	26.8	7.3	No separated data about Ca^{++} and Mg^{++}
Range	7.2–8.0	9.9–11.7	24.0–29.9	7.0–7.5	

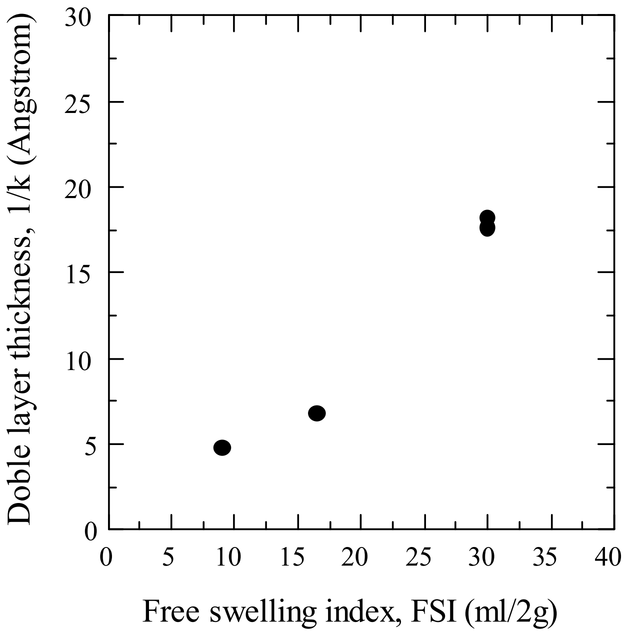


Fig. 5 Relationship between $1/\kappa$ and FSI

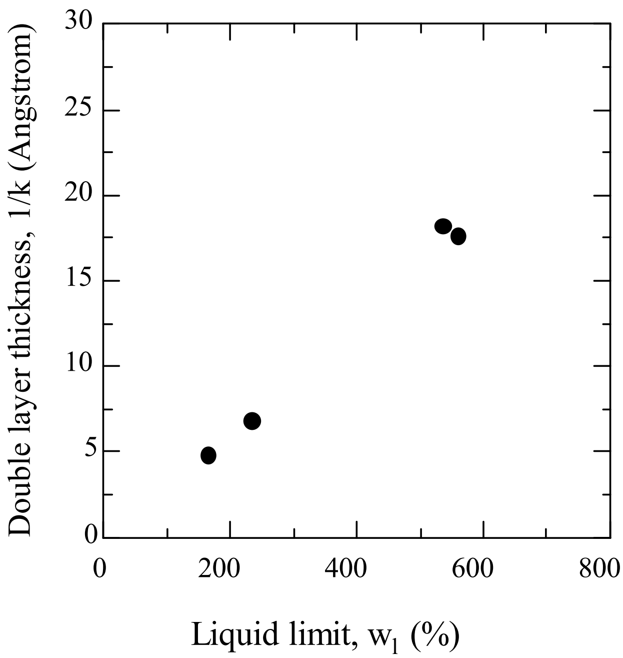


Fig. 6 Relationship between $1/\kappa$ and w_l

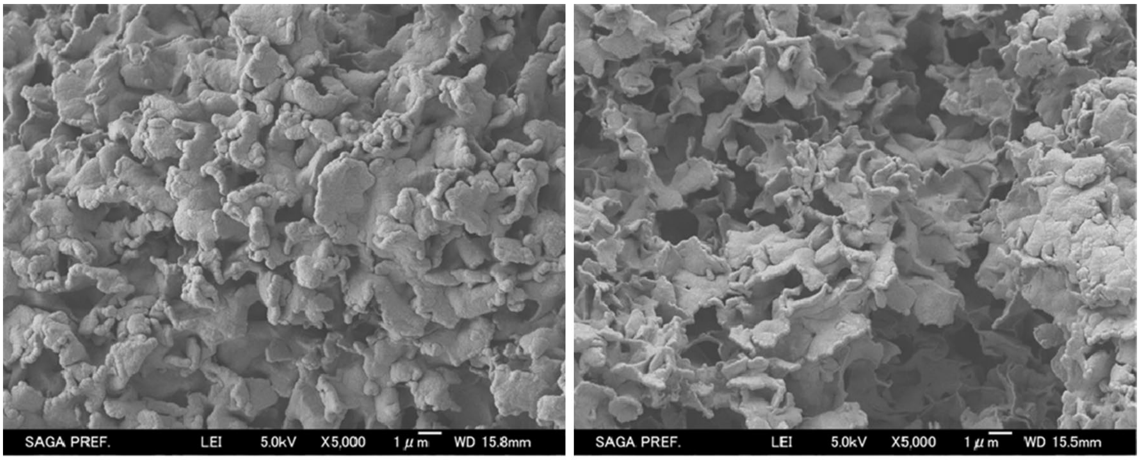
relationships can be observed. Using Eq. (6) and assuming $d = 1/\kappa$, the water contents corresponding to the water in the estimated DDL are also listed in Table 4. The values are about 1/5 of the corresponding w_l. As a reference, if converting the values of FSI listed in Table 1 to water content, on average they are about 2.8 times the corresponding liquid limits.

Swelling Pressure

The value of p_N in Eq. (8) can be considered as the theoretical swelling pressure between two parallel bentonite particles. However, for a mass of bentonite, all the bentonite particles will not be aligned in parallel forms. Using the bentonite sample consolidated in an odometer under 100 kPa with an initial water content of about 300%, scanning electric microscopic (SEM) images are shown in Fig. 7. The horizontal plane is parallel to the top surface of the specimen for the odometer test, and the vertical plane is perpendicular to the horizontal plane. It can be seen that the particles are not lying parallel to each other and there are some face-to-edge contacts. Due to this kind of non-ideal particle alignment, the measured swelling pressure, p_m, should be smaller than the theoretical value of p_N calculated using Eq. (8). To evaluate the van der Waals force, $t = 10$ Å [9] and $A = 2.2 \times 10^{-20}$ J have been adopted in Eq. (7) for all four types of liquid tested. By comparing the theoretical values with the measured values, it has been found that $p_m \approx 0.2\ p_N$. Tripathy et al. [12] reported a similar tendency in case the dry density of the bentonite was lower (below 1600 kg/m^3). When the dry density is lower, the bentonite particles may have more freedom to align in a random form, i.e. away from the parallel alignment, and the swelling pressure will be lower than the theoretical value. When bentonite is compressed under a one-dimensional (1D) deformation condition, its platelet particles will be forced to rotate towards a parallel alignment, i.e. closer to the assumption used in the theory. Using 0.2 p_N as the 'calculated' swelling pressure, p_c, both the calculated and the measured relationships between water content and swelling pressure are compared in Figs. 2, 3 and 4 for the cases of the tap water, the 0.17 mol/l NaCl solution, and the 2.17 mol/l ethanol solution respectively. For all the cases, the values of p_c agree reasonably well with the measured values of p_m. In the case of the 0.1 mol/l $CaCl_2$ solution, with a water content of about 100%, the calculated net interparticle force using Eq. (8) is very small (less than 1.0 kPa).

Conclusions

The swelling behavior of a Na^+-bentonite used in geosynthetic clay linears (GCLs) was investigated using the results of free swelling index (FSI) and swelling pressure tests, as well as liquid limit and plastic limit tests with four types of liquid. The bentonite samples tested had relatively low dry unit weights of 7.45–10.78 kN/m^3. The DDL theory was used for predicting the swelling pressure of the bentonite.



(a) Horizontal plane **(b)** Vertical plane

Fig. 7 SEM image of bentonite

By comparing the calculated and the measured values, the following findings are obtained.

(1) The measured swelling pressures were about 20% of that predicted by DDL theory.
(2) The relationships between the calculated double layer thicknesses ($1/\kappa$) and the measured FSIs and liquid limits of the bentonite are almost linear.
(3) For the case investigated, the calculated water content using the water in the estimated double layer is about 1/5 of the corresponding liquid limit; and if converting the value of FSI to water content, on average it is about 2.8 times the corresponding liquid limit.

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