Evaluation of swelling characteristics of buffer and backfill materials considering the exchangeable-cations compositions of bentonite and its applicability

Évaluation des caractéristiques de gonflement des matériaux tampons et remblais avec prise en compte de compositions échangeuses de cations de la bentonite et de son applicabilité

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ABSTRACT: Bentonite is currently planned to be used as buffer and backfill materials for repositories of high-level nuclear waste, because these materials must have the swelling characteristics to seal the waste. In the design for buffer and backfill materials, we must evaluate quantitatively the swelling behavior of the materials. To solve the problem, this study proposes a new method for predicting the swelling characteristics of buffer and backfill materials. It can consider the influences of the exchangeable-cation compositions of bentonite. This study also demonstrates the applicability of the new prediction method by comparing the predicted results with the experimental data of sand-bentonite mixtures and various kinds of bentonite produced in Japan and the United States.

RÉSUMÉ: Il est actuellement envisagé d'utiliser la bentonite comme matériaux tampons et remblais pour le stockage des déchets nucléaires de haute activité. La bentonite doit avoir les caractéristiques de gonflement servant à enrober les déchets d'une manière étanche. Dans le calcul des matériaux tampons et remblais, on doit estimer d'une façon quantitative leur comportement de gonflement. Cette étude propose une nouvelle méthode permettant de prévoir leur caractéristiques de gonflement à partir de l'effet qu'exercent les composi-tions échangeuses de cations de la bentonite. Cette étude présente également l'applicabilité de la nouvelle méthode en comparant les résultats envisagés avec les données expérimentales sur la mélange sable-bentonite et différents types de bentonites de provenance ja-ponaise et américaine.

1 INTRODUCTION

Compacted bentonite and sand-bentonite mixture are attracting greater attention as buffer and backfill materials for repositories of high-level nuclear waste. These will be used as filling materials of the disposal pits and access tunnels in the underground facilities. Buffer and backfill materials must have the swelling characteristics and are expected to fill up the space between these materials and surrounding ground by swelling. This role is called as "Self-sealing." Figure 1 shows the concept of "Self-sealing."

To design the specifications, such as dry density, sand-bentonite mass ratio and dimensions, of buffer and backfill materials, we must evaluate quantitatively the swelling characteristics of compacted bentonite and sand-bentonite mixtures. In the designs for buffer and backfill materials, we must choose the adequate bentonite among many kinds of bentonite which are produced in the world. From the above-viewpoint, we also have to evaluate quantitatively the swelling behavior of buffer and backfill materials.

From the background described above, this study proposes the new prediction method for swelling characteristics, which can consider the kinds of bentonite and can apply to the sand-bentonite mixtures, to predict the swelling behavior of buffer and backfill materials with high accuracy.

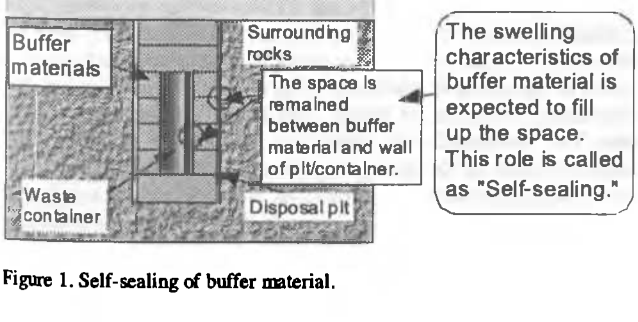


Figure 1. Self-sealing of buffer material.

2 NEW PREDICTION METHOD FOR SWELLING CHARACTERISTICS OF BUFFER AND BACKFILL MATERIALS

To solve the problem described in "Introduction", the author proposed the prediction method for swelling characteristics of compacted bentonite using the theoretical equations of diffuse double layer and the van der Waals force in Komine and Ogata (1994, 1996). This prediction method can consider the influences of chemistry of pore water and temperature, and can be applied to the swelling characteristics of compacted bentonite only. However, it is difficult to apply to the sand-bentonite mixtures because it cannot consider the influences of the mass ratio of sand and bentonite. Moreover, it does not consider the detailed information of the exchangeable-cation composition of bentonite. So, it is also difficult to apply to many kinds of bentonite.

The sands and excavated soils will be used as the materials for buffer and backfill materials from the economical viewpoint in the actual construction (Japan Nuclear Cycle Development Institute 1999, Ogata et al. 1999). In the design for buffer and backfill materials, we also have to choose the adequate bentonite among many kinds of bentonite which are produced in the world. New prediction method for swelling characteristics, which can evaluate the exchangeable-cation compositions in detail and the sand-bentonite mass ratio, is therefore necessary to predict the swelling behavior of the sand-bentonite mixtures and foreign bentonites with high accuracy.

The new prediction method proposed in this study calculates the pressure acting between two montmorillonite layers by weighted averages of repulsive and attractive forces, which are caused by Na, Ca, K, and Mg ions, using the cation-exchange capacities of each ion. Figure 2 shows the concept of the above calculation. The prediction method consists of the theoretical equations for repulsive and attractive forces between two montmorillonite mineral layers which is swelling clay mineral in ben-

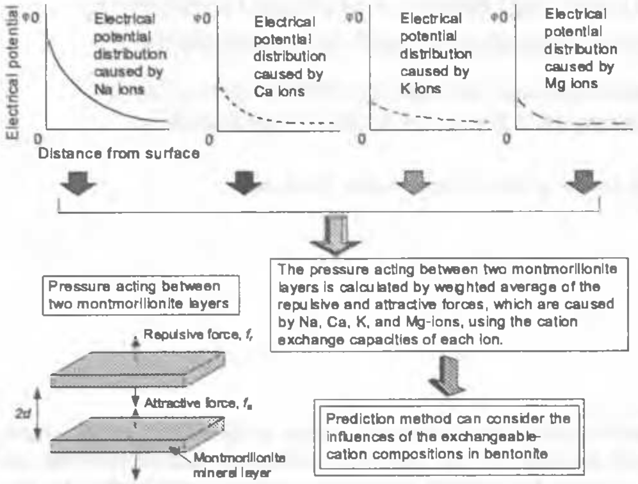


Figure 2. Consideration of exchangeable-cation compositions of bentonite.

tonite, the equations for evaluating the swelling deformation of montmorillonite minerals, and the equations for evaluating the relationship between the swelling deformation of montmorillonite minerals and that of buffer and backfill materials. The proposed method uses the theoretical equations of the Gouy-Chapman diffuse double layer theory and the van der Waals force for evaluating the repulsive and attractive forces of montmorillonite minerals (e.g. Mitchell 1993).

The author proposed the equations for evaluating the swelling behavior of compacted bentonite and sand-bentonite mixtures, which can consider the influences of sand-bentonite mass ratio, ion concentration of pore-water chemistry, specific surface of bentonite, and the exchangeable-cation compositions in bentonite, which significantly influence the swelling characteristics of buffer and backfill materials.

The new prediction method for swelling characteristics of buffer and backfill materials is as follows.

$$p = \frac{1}{CEC} \sum_{\substack{i=Na^+,Ca^{2+} \\ K^+,Mg^{2+}}} \left[EXC_i \left\{ (f_r)_i - (f_a)_i \right\} \right] \tag{1}$$

$$(f_r)_i = 2nkT(\cosh u_i - 1) \times 10^{-3} \tag{1-a}$$

$$u_i = 8\tanh^{-1}\left[\exp(-\kappa_i d_i) \tanh\left(\frac{z_i}{4} \right) \right] \tag{1-b}$$

$$\kappa_i = \sqrt{\frac{2n\upsilon_i^{\,2} e'^2}{\varepsilon kT}} \tag{1-c}$$

$$z_i = 2\sinh^{-1}\left(96.5 \times \frac{EXC_i}{S} \sqrt{\frac{1}{8\varepsilon nkT}} \right) \tag{1-d}$$

$$(f_a)_i = \frac{A_h}{24\pi} \left[\frac{1}{d_i^{\,3}} + \frac{1}{(d_i + t)^3} - \frac{2}{\left(d_i + t/2 \right)^3} \right] \times 10^{-3} \tag{1-e}$$

$$\varepsilon_{sv}^{\,*} = \left\{ e_o + \frac{\varepsilon_{smax}}{100} (e_o + 1) \right\} \times \left\{ 1 + \left(\frac{100}{C_m} - 1 \right) \frac{\rho_m}{\rho_{nm}} + \left(\frac{100}{\alpha} - 1 \right) \frac{100}{C_m} \frac{\rho_m}{\rho_{sand}} \right\} 100 \tag{1-f}$$

$$e_0 = \frac{\rho_{solid}}{\rho_{d0}} - 1 \tag{1-g}$$

$$\rho_{solid} = \frac{\dfrac{100}{C_m} \dfrac{100}{\alpha} \rho_m}{\left\{ 1 + \left(\dfrac{100}{C_m} - 1 \right) \dfrac{\rho_m}{\rho_{nm}} + \left(\dfrac{100}{\alpha} - 1 \right) \dfrac{100}{C_m} \dfrac{\rho_m}{\rho_{sand}} \right\}} \tag{1-h}$$

$$d_i = \frac{\varepsilon_{sv}^{\,*}}{100} \left\{ t + (R_{ion})_i \right\} + (R_{ion})_i \tag{1-i}$$

$$n = \frac{n_0 (\mathrm{mol/m^3}) \times N_A}{1 + \dfrac{\varepsilon_{sv}^{\,*}}{100}} \tag{1-j}$$

$$S = \frac{C_m}{100} S_m + \left(1 - \frac{C_m}{100} \right) S_{nm} \tag{1-k}$$

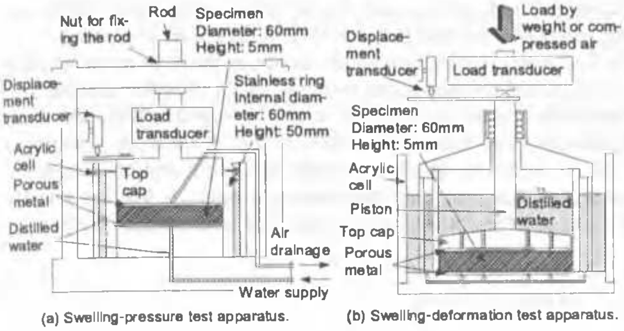
where p is the pressure (kPa) of buffer and backfill materials, $(f_r)_i$ is the repulsive pressure (kPa) between two parallel montmorillonite layers which is caused by the exchangeable-cation i (i denotes someone of Na^+, Ca^{2+}, K^+, and Mg^{2+}), $(f_a)_i$ is the attractive pressure between two parallel montmorillonite layers which is caused by the exchangeable-cation i, EXC_i is the exchange capacity of i (meq/g), CEC is the cation exchange capacity (meq/g), d_i is the half distance between two montmorillonite layers (m) at the exchangeable-cation i, υ_i is the ionic valence of the exchangeable-cation i, e' is the electronic charge (C), k is the Boltzmann constant (J/K), T is the absolute temperature (K), n_0 is the ion concentration ($\mathrm{mol/m^3}$) of pore water, ε is the static permittivity of pore water, A_h is the Hamaker constant (J), t is the thickness of montmorillonite layer (m), $\varepsilon_{sv}^{\,*}$ is the parameter "swelling volumetric strain (%) of montmorillonite" proposed in Komine and Ogata (1999), ε_{smax} is the maximum swelling strain (%) of buffer and backfill materials, e_0 is the initial void ratio of the materials, C_m is the content (%) of montmorillonite in the bentonite by percentage mass, ρ_{d0} is the initial dry density ($\mathrm{Mg/m^3}$), α is the content (%) of bentonite in the materials by percentage mass, ρ_m is the particle density ($\mathrm{Mg/m^3}$) of montmorillonite, ρ_{nm} is the particle density ($\mathrm{Mg/m^3}$) of component minerals excluding montmorillonite in the bentonite, ρ_{sand} is the particle density ($\mathrm{Mg/m^3}$) of sand in the materials, $(R_{ion})_i$ is the nonhydrated radius of the exchangeable-cation i (m), N_A is Avogadro's number, S is the specific surface ($\mathrm{m^2/g}$) of bentonite, S_m is the specific surface ($\mathrm{m^2/g}$) of montmorillonite and S_{nm} is the specific surface ($\mathrm{m^2/g}$) of component minerals excluding montmorillonite in the bentonite.

3 APPLICABILITY OF NEW PREDICTION

This section describes the applicability of prediction method proposed in this study by comparing the predicted results with experimental results of various bentonite and sand-bentonite mixtures (Komine and Ogata 1996, 1997, 1999)

3.1 *Sample and experiment*

This study used four kinds of commercial bentonite. Bentonite A, which is called Kunigel-V1, is produced at the Tsukinuno Mine in Yamagata prefecture of Japan. This is sodium-type bentonite containing nearly 48% montmorillonite, and is frequently used in the studies on the material for artificial barriers against nuclear waste in Japan. Bentonite B called as Volclay is produced at the Wyoming in the United States. This is also sodium-type bentonite. The montmorillonite content is nearly 69%. Bentonite C, which is called as Kunibond, is produced at the Dobuyama Mine in Miyagi prefecture of Japan. This is calcium-type bentonite. The montmorillonite content is nearly 80%. Bentonite D called as Neokunibond is produced at Kawasaki-cho Mine in Miyagi prefecture of Japan. The montmorillonite content is nearly 76%.



(a) Swelling-pressure test apparatus. (b) Swelling-deformation test apparatus.

Figure 3. Schematic drawing of test apparatuses.

This is artificial sodium-type bentonite, to which the calcium-type bentonite is transformed by sodium carbonate solution. The montmorillonite content was calculated by the methylene blue absorption values of each bentonite and montmorillonite (White and Michael, 1979). The methylene blue absorption values of Bentonite A, B, C and D are 67, 94, 112 and 106 meq/100g, respectively. That of montmorillonite is 140 meq/100g. The sand-bentonite mixtures were produced by mixing Bentonite A and Mikawa silicate sand No. 6.

Test apparatuses are shown in Fig. 3. The swelling-pressure test measured the swelling pressure of the materials as water was supplied from the bottom while the specimen was confined. In the swelling-deformation test, the relationship between the axial swelling-deformation and the time required from the start of water supply was measured. The experimental procedures were described in Komine and Ogata (1996, 1999).

3.2 *Parameters and predictions*

Tables 1 and 2 show the parameters for prediction. Table 1 shows the parameters which are dependent on the kinds of bentonite. Table 2 shows the other parameters which are independent on the kinds of bentonite.

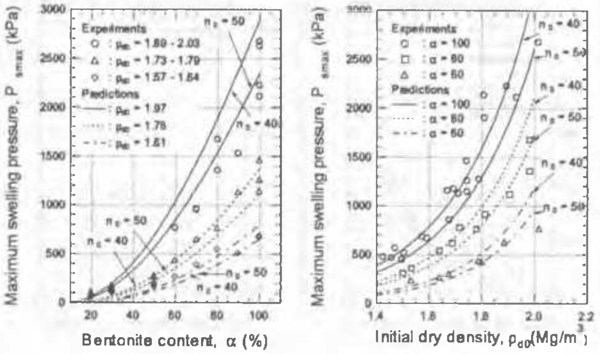
The parameters shown in Table 1 were measured values of each bentonite. The particle density of sand shown in Table 2 is the value of Mikawa silicate sand No. 6 which was used in the experiments of Komine and Ogata (1999). The absolute temperature T is 295 K because the laboratory is maintained at constant temperature of 22 in the centigrade scale. The other parameters are quoted from Mitchell (1993), and Komine and Ogata (1996).

Table 1. Parameters which are dependent on the kinds of bentonite.

Bentonite	A	B	C	D
ρ_m (Mg/m^3)	2.77	2.77	2.77	2.77
ρ_{nm} (Mg/m^3)	2.81	3.01	2.50	2.43
C_m (%)	48	69	80	76
CEC (meq/100g)	0.732	1.007	0.796	1.035
EXC_{Na^+} (meq/100g)	0.405	0.566	0.119	0.620
$EXC_{Ca^{2+}}$ (meq/100g)	0.287	0.293	0.585	0.333
EXC_{K^+} (meq/100g)	0.009	0.016	0.019	0.019
$EXC_{Mg^{2+}}$ (meq/100g)	0.030	0.132	0.072	0.063

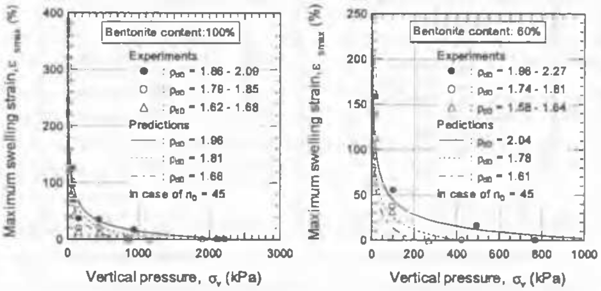
Table 2. Other parameters.

ρ_{sand} (Mg/m^3)	2.66
S_m (m^2/g)	810
S_{nm} (m^2/g)	0
$(R_{ion})_{Na}$ (nm)	0.098
$(R_{ion})_{Ca}$ (nm)	0.1115
$(R_{ion})_K$ (nm)	0.133
$(R_{ion})_{Mg}$ (nm)	0.0835
υ_{Na}	1
υ_{Ca}	2
υ_K	1
υ_{Mg}	2
t (m)	9.60×10^{-10}
e' (C)	1.602×10^{-19}
Boltzmann constant k (J/K)	1.38×10^{-23}
Hamaker constant A_h (J)	2.2×10^{-20}
Avogadro's Number N_A	6.023×10^{23}
ε (C^2/(Jm))	$80 \times 8.8542 \times 10^{-12}$
T (K)	295



Notes: n_0=Ion concentration of pore water (mol/m^3)

Figure 4. Swelling-pressure of the mixtures of sand and bentonite.



Notes: ρ_{d0}=Initial dry density (Mg/m^3), n_0=Ion concentration of pore water (mol/m^3)

Figure 5. Swelling-deformation of the mixtures of sand and bentonite.

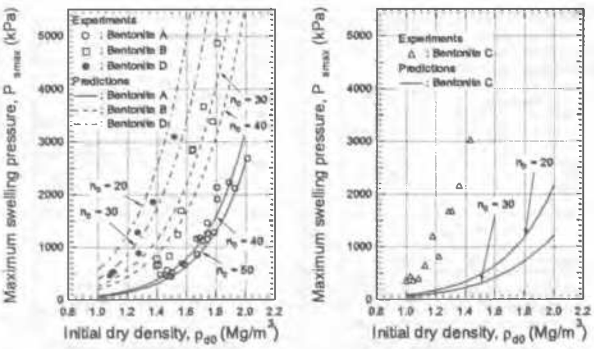
Figures 4 and 5 show the comparisons of predicted results with experimental results for swelling characteristics of the mixtures of Bentonite A and Mikawa silicate sand No. 6.

Figure 4 indicates that the maximum swelling pressure is strongly dependent on the bentonite content and the initial dry density. The maximum swelling pressure increases as the bentonite content increases at the same initial dry density. At the same bentonite content, the maximum swelling pressure also increases as the initial dry density increases. The same relationships calculated by the proposed prediction are shown by the curves in Fig. 4. The maximum swelling pressure is equal to the pressure p calculated by Eq. (1) in the prediction.

Figure 5 shows the experimental results of swelling-deformation tests at the bentonite content of 60 and 100%. In Fig. 5, the maximum swelling pressure measured with the swelling deformation restricted corresponds to the vertical pressure at $\varepsilon_{smax} = 0$. The curves in Fig. 5 show the relationships between the maximum swelling strain and vertical pressure calculated by the prediction proposed. In these calculations, the vertical pressure is equal to the pressure p calculated by Eq. (1). The curves calculated from the prediction closely agree with the experimental results as shown in Figs 4 and 5.

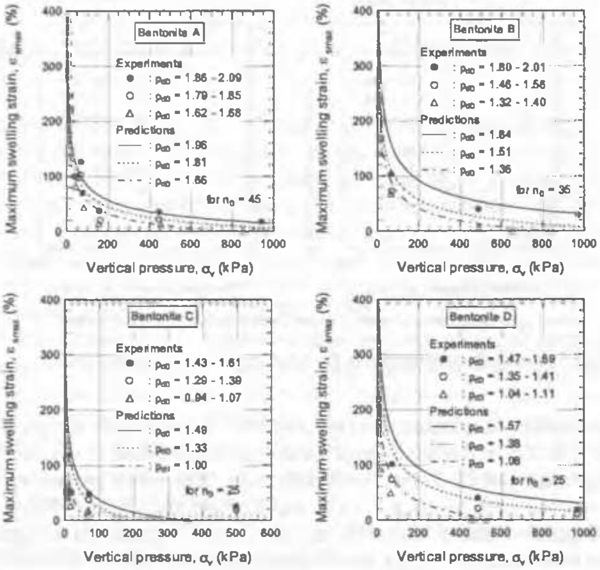
Figures 6 and 7 show the comparisons of predicted results with experimental results for swelling characteristics of various bentonite.

Figure 6 shows the relationships between the maximum swelling pressure and the initial dry density for each bentonite. The same relationships calculated by the proposed prediction are also shown by the curves in Fig. 6. This figure indicates that the predicted results have good agreement with the experimental results for Bentonite A, B, and D. Whereas, the calculated maximum swelling pressure of Bentonite C, which is the calcium type, is lower than the experimental results. This tendency is stronger as the initial dry density increases.



Notes: n_0=Ion concentration of pore water (mol/m^3)

Figure 6. Swelling-pressure of various bentonites.



Notes1: In case of Bentonite C, the predicted results have good agreement with the experimental results on the condition that the vertical pressure is less than 100kPa. If the vertical pressure is more than 100kPa, the accuracy of prediction is not satisfactory.

Notes2: ρ_{d0}=Initial dry density (Mg/m^3), n_0=Ion concentration of pore water (mol/m^3)

Figure 7. Swelling-deformation of various bentonites.

In the theoretical equations (1-a) – (1-d) of the Gouy-Chapman diffuse double layer theory, it is assumed that the cations are point charges, whereas, ions are of finite size (Mitchell 1993). Moreover, Sposito (1989) indicated that the diffuse double layer theory might not give accurate predictions for bivalent electrolytes, though the theory can give accurate predictions for monovalent electrolytes. From the above considerations, the applicability of the diffuse double layer theory declines for evaluating the swelling properties of the calcium-type bentonite such as Bentonite C.

Figure 7 shows the comparisons of the predicted results with the experimental results for swelling-deformation tests. The curves calculated from the prediction closely agree with the experimental results shown in Fig. 7 in the cases of Bentonite A, B, and D, of which the main exchangeable-cation is the sodium-ion.

It is also found that the predicted results for Bentonite C have good agreement with the experimental results on condition that the vertical pressure is less than 100 kPa from Fig. 7. However, the accuracy of prediction is not satisfactory if the vertical pressure is more than 100 kPa. The above is also caused by the decline of applying the diffuse double layer theory to evaluate the swelling properties of the calcium-type bentonite.

Therefore, the prediction method proposed in this study can predict adequately the swelling characteristics of sodium-type bentonite. In the projects for disposing the radioactive wastes in some countries, the sodium-type bentonite is planned to be used (Atomic Energy of Canada Limited 1994, Ogata et al. 1999, Swedish Nuclear Fuel and Waste Management Co. 1992). So, the prediction method proposed will be available in practice.

The new prediction method can obtain the maximum swelling pressure and the maximum swelling strain of buffer and backfill materials at various kinds of bentonite, dry densities and sand-bentonite mass ratios. Therefore, it can be used for selecting a kind of bentonite which is suitable for buffer and backfill materials, and for designing the bentonite content, the compaction density and the dimension from the viewpoint of "Self-sealing."

4 CONCLUSIONS

This study proposed the new method for predicting the swelling characteristics of buffer and backfill materials. It can consider the influences of the exchangeable-cation compositions of bentonite and the sand-bentonite mass ratio. Furthermore, this study clarifies the applicability of the new prediction method by comparing the predicted results with the experimental data of sand-bentonite mixtures and various kinds of bentonite produced in Japan and the United States.

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