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Theoretical equations for evaluating hydraulic conductivities of bentonite based buffer and backfill

Équations théoriques pour des conductivités hydrauliques d'évaluation de bentonite basée sur des matériaux tampons et des matériaux de colmatage en revers

H. Komine
Ibaraki University, Department of Urban and Civil Engineering, Japan

ABSTRACT
Theoretical new equations for evaluating the hydraulic conductivity of compacted bentonites and bentonite-sand mixtures are derived for designing bentonite based buffer and backfill materials to fill high-level nuclear waste repositories. New equations for evaluating the velocity of interlayer-water flow between two montmorillonite layers considering the swelling behaviors of montmorillonite are proposed. Furthermore, a calculating method for the hydraulic conductivity of compacted bentonite and bentonite-sand mixtures is presented by combining the new equations with the equations for evaluating swelling behavior of montmorillonite in bentonite, which have already been proposed by the author. The applicability of this method was investigated by comparing the calculated results with laboratory test results on the hydraulic conductivity of compacted bentonites and bentonite-sand mixtures.

RÉSUMÉ
Les nouvelles équations théoriques pour des conductivités hydrauliques d'évaluation de bentonite compacte et de mélanges bentonite-sable ont été trouvées pour désigner de la bentonite basée sur des matériaux tampons et des matériaux de colmatage en revers, afin de combler le haut niveau des entrepôts de déchets nucléaires. Nous proposons de nouvelles équations pour évaluer le taux de débit d'eau situé entre les couches, entre deux couches de montmorillonite, afin d'examiner la manière dont la montmorillonite gonfle. De plus, nous présentons une méthode prédictive sur les conductivités hydrauliques de bentonite compacte et de mélanges bentonite-sable, en associant les nouvelles équations avec les équations qui évaluent la manière dont la montmorillonite gonfle dans la bentonite, équations qui ont déjà été présentées par l'auteur. L'applicabilité de cette méthode a été examinée afin de comparer les résultats prévus avec les résultats des tests de laboratoires au sujet des conductivités hydrauliques de bentonite compacte et de mélanges bentonite-sable.

1 INTRODUCTION

Design of bentonite based buffer and backfill materials to fill disposal facilities are important for developing technology for high-level nuclear wastes disposal. A disposal facility at a depth of several hundred meters is currently planned in Japan. Figure 1 shows an example of disposal facility proposed in the Japanese program (Ogata et al., 1999). The function of buffer material is to create a very low permeability zone around the containers since high-level radioactive wastes must be kept separate from the surrounding environment for an extended time. The function of backfill material is also to create a low permeable zone in the access tunnel of underground disposal facility. From the above background, compacted bentonite and sand-bentonite mixture are attracting greater attention as buffer and backfill materials because they have very low permeable.

To design the specifications, such as dry density and sand-bentonite mass ratio, of buffer and backfill materials, we must investigate the hydraulic properties by experiments and evaluate quantitatively the hydraulic conductivities of compacted bentonite and sand-bentonite mixtures.

The author has investigated the hydraulic conductivities of bentonite based materials by experimental works in Komine (2003; 2004). The experimental works for obtaining hydraulic conductivities of bentonite based buffer and backfill materials are required very long terms such as more than 1 year. Therefore, it is not realistic to measure the hydraulic conductivities of buffer and backfill materials which are made of many kinds of bentonite produced in the world because very long terms are needed. From the viewpoint of very low permeability, we must evaluate quantitatively the hydraulic conductivities of compacted bentonite and sand-bentonite mixtures in the design. To satisfy above-mentioned objective, this study proposes a calculating method for hydraulic conductivities of compacted bentonite and sand-bentonite mixtures.

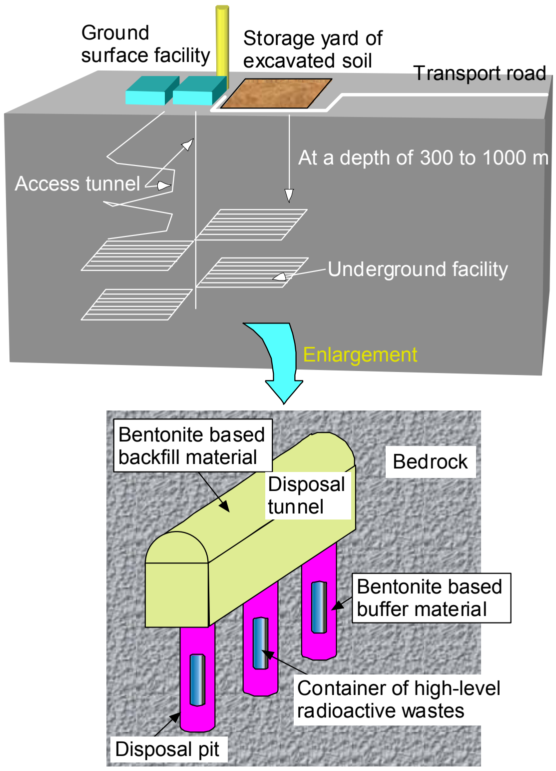

Figure 1. An example of disposal facility proposed in Ogata et al.(1999)

2 THEORETICAL EQUATIONS FOR HYDRAULIC CONDUCTIVITY OF BENTONITE BASED BUFFER AND BACKFILL MATERIALS

The theoretical equations proposed in this study for hydraulic conductivity comprises some equations that are newly proposed for streamline-flow model between two montmorillonite layers, and were previously developed equations for the swelling volumetric strain of montmorillonite.

2.1 *Swelling volumetric strain of montmorillonite*

Komine and Ogata (1999; 2004) have proposed the parameter "Swelling volumetric strain of montmorillonite, ε_{sv}^{*} (%)" to evaluate quantitatively the physical properties such as swelling and hydraulic conductivity of bentonite based buffer and backfill materials.

They also observed the swelling behavior of bentonite by using scanning electron microscope which can control the temperature and vapor pressure around samples. From the observed results, it was found that the voids in the material were filled completely by swelling deformations of bentonite absorbing water. Furthermore, they discussed the swelling processes of bentonite based buffer and backfill materials as shown in Fig. 2. Equations (1) – (3) of the parameter ε_{sv}^{*} are derived from the above discussions. Komine and Ogata (1996; 1999) discuss the details of swelling processes and the derivations of Eqs. (1), (2) and (3).

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

$$e_0 = \frac{\rho_{solid}}{\rho_{d0}} - 1 \tag{2}$$

$$\rho_{solid} = \frac{\dfrac{100}{C_m}\dfrac{100}{\alpha}\rho_m}{\left\{1 + \left(\dfrac{100}{C_m} - 1\right)\dfrac{\rho_m}{\rho_{nm}} + \left(\dfrac{100}{\alpha} - 1\right)\dfrac{100}{C_m}\dfrac{\rho_m}{\rho_{sand}}\right\}} \tag{3}$$

where ε_{smax} is the maximum swelling strain (%) of buffer and backfill materials, e_0 is the void ratio of the materials, C_m is the content (%) of montmorillonite in the bentonite by percentage mass, ρ_{d0} is the dry density (Mg/m^3) of the materials, α is the content (%) of bentonite in the materials by percentage mass, ρ_m is the particle density (Mg/m^3) of montmorillonite, ρ_{nm} is the particle density (Mg/m^3) of component minerals excluding montmorillonite in the bentonite, and ρ_{sand} is the particle density (Mg/m^3) of sand in the materials.

The parameter ε_{sv}^{*} can be also evaluated from the swelling deformation of a montmorillonite mineral. A model-simulated deformation of a montmorillonite mineral was produced, and an equation was proposed for evaluating the relationship between the parameter ε_{sv}^{*} and the distance $2d$ between two parallel montmorillonite layers in Komine and Ogata (1996). Furthermore, Komine and Ogata (2003; 2004) proposed the developed equation (4) which can accommodate the influences of exchangeable-cation composition of bentonite by the parameters concerning the numbers of Na^{+}, Ca^{2+}, K^{+} and Mg^{2+}, and the diameter of exchangeable-cations.

$$d_i = \frac{\varepsilon_{sv}^{*}}{100}\left\{t + \left(R_{ion}\right)_i\right\} + \left(R_{ion}\right)_i \tag{4}$$

where d_i is the half distance between two montmorillonite layers (m) at the exchangeable-cation i (i denotes either of Na^{+}, Ca^{2+}, K^{+}, and Mg^{2+}), t is the thickness of the montmorillonite layer (m), and $(R_{ion})_i$ is the nonhydrated radius of the exchangeable-cation i (m).

Eqs. (1) – (4) can be used to evaluate the relationship between the maximum swelling strain of buffer and backfill material and half the distance between two parallel montmorillonite layers through the intermediary of the parameter ε_{sv}^{*}.

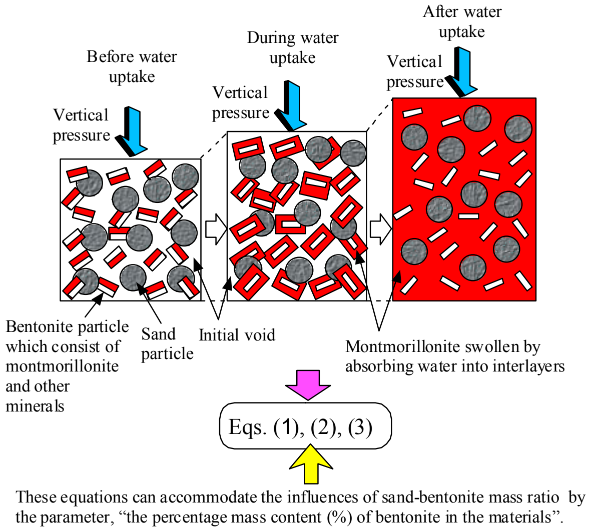

Figure 2. Swelling processes of bentonite based buffer and backfill.

2.2 *Flow model between two montmorillonite layers*

From the observed results of swelling behavior of bentonite in Komine and Ogata (1999; 2004), the voids in the bentonite based materials were filled completely by swelling deformations of bentonite absorbing water. Therefore, it is assumed that water goes mainly through between two montmorillonite layers swollen by absorbing water as shown in Fig. 3. Furthermore, it is also assumed that the velocity of water flow between two montmorillonite layers dominates the velocity of water flow in the bentonite based material because it is conceivable that the water flow between two layers is the minimum in the material. From the above discussion, hydraulic conductivity of bentonite based buffer and backfill materials can be predicted by calculating the velocity of water flow between two montmorillonite layers.

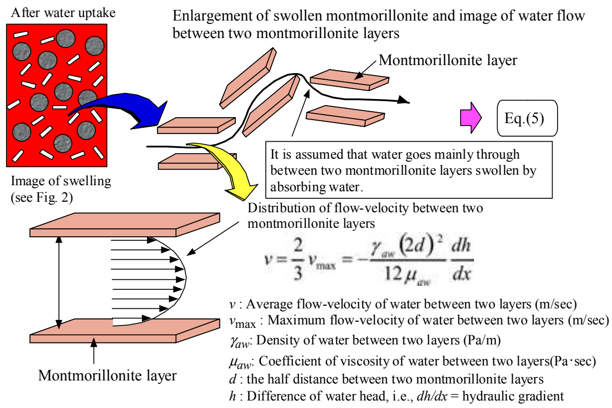

Figure 3. Image of main pathway of water flow in bentonite based buffer and backfill materials after absorbing water.

Figure 3 also shows the streamline-flow model between two montmorillonite parallel-plate layers. In this model, it is assumed that water flow is two-dimensional and water is incompressible. This model is equivalent of Poiseuille flow and the hydraulic conductivity of two montmorillonite parallel-plate layer can be calculated by,

$$k_i = \frac{\gamma_{aw}}{12\,\mu_{aw}}(2d_i)^2 \tag{5}$$

where k_i is the hydraulic conductivity (m/sec) of two montmorillonite parallel-plate layers at the exchangeable-cation i, γ_{aw} is the density of interlayer water between two montmorillonite parallel-plate layers (Pa/m), and μ_{aw} is the coefficient of viscosity of interlayer water between two montmorillonite parallel-plate layers ($\mathrm{Pa \cdot sec}$).

The above equation (5) also can accommodate the influences of exchangeable-cation composition of bentonite by the parameters concerning the numbers of Na^+, Ca^{2+}, K^+ and Mg^{2+}, and the diameter of exchangeable-cations.

Consequently, the hydraulic conductivity of bentonite based buffer and backfill materials can be calculated by,

$$k = \frac{1}{CEC} \sum_{\substack{i=Na^+,Ca^{2+}\\K^+,Mg^{2+}}} \left[EXC_i\, k_i\right] \tag{6}$$

where k is the hydraulic conductivity (m/sec) of bentonite based buffer and backfill materials, EXC_i is the exchange capacity of the exchangeable-cation i (meq/g) and CEC is the cation exchange capacity of bentonite (meq/g).

Hydraulic conductivity of bentonite based buffer and backfill materials can be evaluated by combining Eqs. (1) – (6).

3 APPLICABILITY OF THEORETICAL EQUATIONS FOR HYDRAULIC CONDUCTIVITY OF BUFFER AND BACKFILL MATERIALS

In the section, the applicability of the theoretical equations has been confirmed by comparing the calculated results with experimental results in Komine (2003; 2004) on the hydraulic conductivities of compacted bentonites and sand-bentonite mixtures.

3.1 *Parameters and physical constants in the theoretical equations for hydraulic conductivity*

This section describes the parameters and physical constants for the preceding theoretical equations. Table 1 shows the parameters determined by the fundamental characteristics of bentonite and sand.

Table 1. Parameters and physical constants.

ρ_m	2.77 Mg/m^3
ρ_{nm}	2.81 Mg/m^3
ρ_{sand}	2.66 Mg/m^3
C_m	48 %
CEC	0.732 meq/g
$EXC_{Na}{}^+$	0.405 meq/g
$EXC_{Ca}{}^{2+}$	0.287 meq/g
$EXC_K{}^+$	0.009 meq/g
$EXC_{Mg}{}^{2+}$	0.030 meq/g
$(R_{ion})_{Na}$	0.098 nm
$(R_{ion})_{Ca}$	0.1115 nm
$(R_{ion})_K$	0.133 nm
$(R_{ion})_{Mg}$	0.0835 nm
t	9.60×10^{-10} m

The parameters shown in Table 1 are determined on the basis of the values quoted from the previous papers and the measured values of Kunigel-V1 and Mikawa silicate sand No. 6, which are the materials used in Komine (2004). The values of ρ_m and ρ_{nm} in Table 1 are quoted from Komine and Ogata (2003). ρ_{sand} is the measured value of Mikawa silicate sand No. 6. The montmorillonite content C_m is 48 %. It is calculated at the ratio of methylene blue absorption values of bentonite and montmorillonite. $EXC_{Na}{}^+$, $EXC_{Ca}{}^{2+}$, $EXC_K{}^+$, and $EXC_{Mg}{}^{2+}$ are also measured values of Kunigel-V1. They are measured values of the eduction by one-normal CH_3COONH_4 solution. The chemical analysis of the eduction was done by the ICP-atomic emission spectrometer. *CEC* is the sum of the above values.

The nonhydrated radius of the exchangeable-cation i $(R_{ion})_i$ is also quoted from Komine and Ogata (2003). The value of $(R_{ion})_i$ shown in Table 1 is determined by referring to the values of nonhydrated radius of Na^+, Ca^{2+}, K^+ and Mg^{2+}. The value of thickness of montmorillonite layer t is quoted from Grim (1968).

It is well-known that the density and the coefficient of viscosity of water depend on the temperature of circumstance. Therefore, the density and the coefficient of viscosity of free water in soil materials such as bentonite can be calculated by the equations (7) and (8). Equation (7) is the regression curves between density of water and temperature, and equation (8) can be obtained by regressing the relationship between coefficient of viscosity of free water and temperature. These equations can obtain the density of water and the coefficient of viscosity of free water according to temperature.

$$\gamma_{fw} = 8512.9 + 1295.2 \times \exp\left[-\left\{\frac{(T-273)+0.4946}{163.45}\right\}^2\right] \tag{7}$$

$$\mu_{fw} = 0.00026087 + 0.0015058 \times \exp\left[-0.034688 \times (T-273)\right] \tag{8}$$

where γ_{fw} is the density of free water in the materials (Pa/m), and μ_{fw} is the coefficient of viscosity of free water in the materials ($\mathrm{Pa \cdot sec}$). T is the absolute temperature (K).

The interlayer water between montmorillonite mineral layers contains the large number of cations such as Na^+, Ca^{2+}, K^+, Mg^{2+}, etc. These cations hydrate and are absorbed to the vicinity of surface of montmorillonite mineral. A water molecule is dipolar, so is also absorbed electrochemically to surface of montmorillonite mineral which is negative charge. Furthermore, hydrogen bond is generated between oxygen molecule of montmorillonite mineral and water molecule.

As described above, the interlayer water between montmorillonite mineral layers is strongly absorbed to the surface of montmorillonite mineral, so the density and the coefficient of viscosity of the interlayer water between montmorillonite mineral layers are larger than those of free water. Equation (9) can calculate the ratio of properties between interlayer water and free water.

$$\frac{\gamma_{aw}}{\mu_{aw}} = R\,\frac{\gamma_{fw}}{\mu_{fw}} \tag{9}$$

where γ_{aw} is the density of interlayer water between montmorillonite mineral layers (Pa/m), μ_{aw} is the coefficient of viscosity of interlayer water between montmorillonite mineral layers ($\mathrm{Pa \cdot sec}$), and R is the parameter on ratio of density and coefficient of viscosity between interlayer water and free water.

In this study, the value of parameter, R is quoted from Sato (1971) and/or in Sato and Murota (1971). In these researches, they run the permeability tests of some kinds of soil material such as silt, clay and bentonite on the condition of very low hydraulic-gradient. They obtained the values of R by analyzing the experimental results mentioned above from the viewpoint of hydrodynamics. According to their researches (Sato 1971; Sato and Murota 1971), they proposed R=79 for bentonite clay, and also proposed R=14 for silty clay. Van Olphen (1991) introduces many previous researches about the density and viscosity of absorbed water in the vicinity of clay particles. The above values of R are valid according to these previous researches. Therefore, Table 2 shows the parameters of free water and water between two montmorillonite parallel-plate layers in this study. γ_{fw} and μ_{fw} are calculated by equations (7) and (8) substituting T=295K, respectively.

Consequently, hydraulic conductivity of compacted bentonites and sand-bentonite mixtures can be evaluated by combining Eqs. (1) – (9).

Table 2. Parameters of free water and water between two montmorillonite parallel-plate layers

γ_{fw}	9783.8 Pa/m (It is calculated by Eq. (7) at T=295K.)
μ_{fw}	0.000963 Pa · sec (It is calculated by Eq. (11) at T=295K.)
R	14, 79

3.2 *Comparison of calculated and experimental results*

Figures 4 and 5 show the comparisons of calculated results with experimental results of hydraulic conductivity of bentonite based buffer and backfill materials. Komine (2003; 2004) report the details of experiment and test procedure for measuring the hydraulic conductivity shown in Figs. 4 and 5. Figure 4 shows the compared results of backfill material of which bentonite content is within the range of 5 to 50% and dry density is the range of 1.43 to 1.79 Mg/m^3. Figure 5 shows the relationship between calculated and experimental results of buffer material of which bentonite content is in 50% to 100% and dry density is the range of 1.71 to 2.17 Mg/m^3.

These figures indicate that the calculated results concur well with the experimental results for both of buffer and backfill materials. Especially, the calculation of hydraulic conductivity by combining Eqs. (1) – (9) can predict acceptably the reduction of hydraulic conductivity with the increase of dry density and bentonite content of buffer and backfill materials.

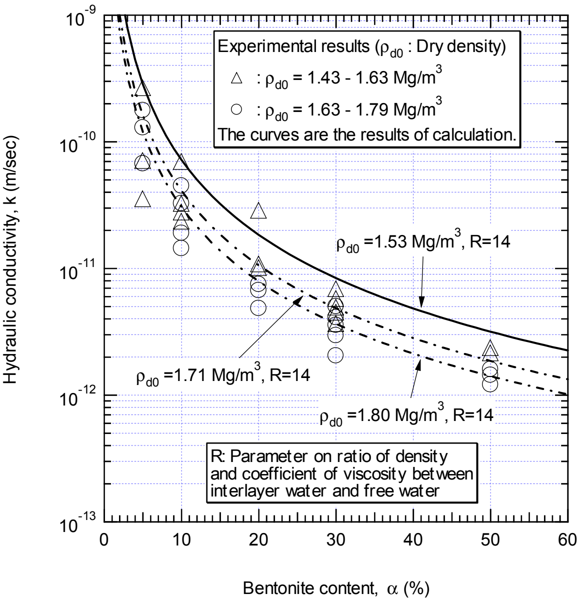

Figure 4. Comparison of calculated results with experimental results (Komine, 2003; 2004) for backfill material.

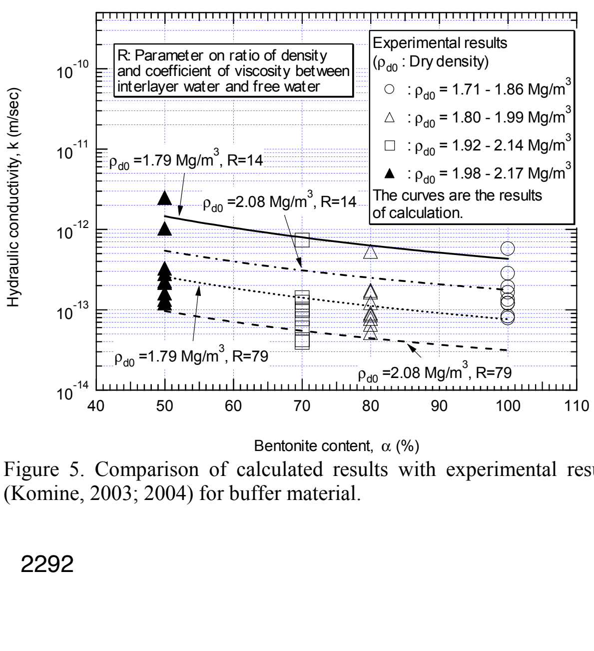

Figure 5. Comparison of calculated results with experimental results (Komine, 2003; 2004) for buffer material.

The theoretical equations of hydraulic conductivity proposed in this study can calculate the hydraulic conductivities of bentonite based buffer and backfill materials at various dry densities and bentonite contents. Therefore, it can be used for designing the bentonite content and compaction density to achieve very low permeability.

4 CONCLUSIONS

This study proposed the theoretical equations for calculating hydraulic conductivity of bentonite based buffer and backfill materials according to bentonite content and dry density. The proposed equations for hydraulic conductivity comprises some equations that are newly proposed for streamline-flow model between two montmorillonite parallel-plate layer, and were previously developed equations for the swelling volumetric strain of montmorillonite (Komine and Ogata, 1999). Furthermore, the applicability of the theoretical equations has been confirmed by comparing the predicted results with laboratory test results (Komine, 2003; 2004) on the hydraulic conductivities of compacted bentonites and sand-bentonite mixtures. From the results. the theoretical equations proposed in this study can calculate the hydraulic conductivity of bentonite based buffer and backfill materials at various dry densities and bentonite contents. It can be used for designing the bentonite content and compaction density of buffer and backfill materials to achieve very low permeability.

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