

## Influence of Water Salinity on the Hydraulic Conductivity of Compacted Bentonite

물의 염도가 압축벤토나이트의 수리전도도에 미치는 영향

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### Abstract

The influence of water salinity on the hydraulic conductivities of compacted bentonites with several dry densities were studied. The hydraulic conductivity increases with increasing salinity only when the dry density of bentonite is relatively low. The degree of increase becomes more remarkable at a lower dry density of bentonite. For bentonite with the density of 1.0 Mg/m<sup>3</sup> and 1.2 Mg/m<sup>3</sup>, the hydraulic conductivity of the 0.4 M NaCl solution increases up to about 7 times and 3 times, respectively higher than that of freshwater. However, for the bentonite with a dry density higher than 1.4 Mg/m<sup>3</sup>, the salinity has an insignificant effect on the hydraulic conductivity, and the hydraulic conductivity is nearly constant within the salinity range of 0.04 to 0.4 M NaCl. The pre-saturation of the bentonite specimen with freshwater has no significant influence on the hydraulic conductivity.

**Key words** : hydraulic conductivity, bentonite, salinity effects, diffuse double layer, electric potential

### 요 약

다양한 건조밀도를 가진 압축벤토나이트의 수리전도도에 물의 염도가 미치는 영향이 조사되었다. 압축벤토나이트의 수리전도도는 벤토나이트의 건조밀도가 상대적으로 낮은 경우에만, 염도가 증가함에 따라 증가하였으며, 염도의 증가에 따른 수리전도도의 증가 정도는 벤토나이트의 건조밀도가 낮을수록 더 현저하였다. 건조밀도가 1.0 Mg/m<sup>3</sup> 및 1.2 Mg/m<sup>3</sup> 인 압축벤토나이트의 경우, 0.4 M NaCl 용액의 수리전도도는 탈염수의 경우에 비해 각각 7배 및 3배가 증가하였다. 그러나 1.4 Mg/m<sup>3</sup> 보다 큰 건조밀도를 가진 압축벤토나이트의 경우에는, 수리전도도에 미치는 염도의 영향이 크지 않았으며, NaCl의 농도가 0.04 M에서 0.4 M 인 범위에서는 거의 일정한 값을 유지하였다. 벤토나이트 시편을 탈염수로 미리 포화시키는 것은 수리전도도에 큰 영향을 미치지 않았다.

**중심단어** : 수리전도도, 벤토나이트, 염도 영향, 확산이중층, 전기포텐셜

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## I. Introduction

A high-level waste repository would be constructed in a crystalline hard rock formation at a depth of several hundred meters below ground surface. In the Korean reference high-level waste disposal system (KRS) [1], the repository would be expected to be of room-and-pillar design, and will be a network of access tunnels and disposal rooms with vertical shafts extending from the surface to the access tunnels (Fig. 1). The spent fuels are packed in the disposal canister that consists of an inner carbon steel vessel and an outer copper shell. The canister would be deposited in an array of large-diameter boreholes drilled on the floors of emplacement rooms, and the gap between the canister and the wall of the borehole would be filled with a buffer material. The buffer is one of the major components of the engineered barriers in the repository, and is required to inhibit the release of radionuclides from the waste to the surrounding rock. In many countries [2-4], the conceptual design for an underground repository in hard rock formation includes the use of compacted bentonite as a buffer material. The KRS will also use the compacted bentonite-based materials as both buffer surrounding waste canister and backfill for tunnels and access shafts (Fig. 2). As such a repository would be located below an aquifer, the buffer would be fully saturated with groundwater sometime after closure of the repository. Because of the low hydraulic conductivity and the high sorption capacity of compacted bentonite, molecular diffusion will be the principal mechanism by which radionuclide will migrate through the buffer.

After closure of the repository located in a coastal area, the salinity of the groundwater in the host rock is expected to be increased due to the seawater intrusion. The increase of the salinity might increase the hydraulic conductivity in the buffer and enhance the water movement through the buffer. The susceptibility of clay to changes in hydraulic conductivity upon exposure to saline water has been an important factor in the design of a containment barrier near a beach and a liner to contain brine. It has been shown by a number of researchers [5-7] that dramatic increase in hydraulic conductivity may occur when a concentrated salt solution is

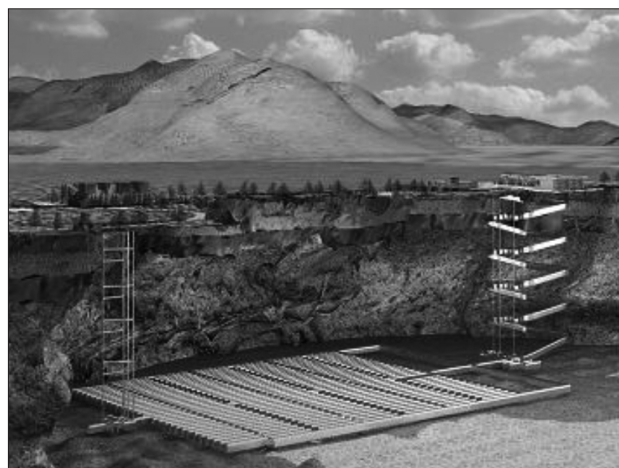


Fig. 1. Cutaway view of the Korean reference high-level waste disposal system.

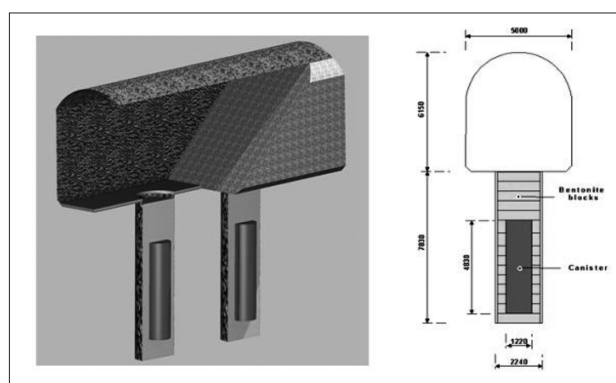


Fig. 2. Cross section of the engineered barrier system of the Korean reference high-level waste disposal system.

introduced into a freshwater clay. However the effects of saline water with relatively low salt concentration on the hydraulic conductivity of compacted clay have not been fully understood yet.

This study intends to investigate the possible effects of low salinity of water on the sealing performance of buffer for the high-level waste repository. The results of experimental studies to investigate the effect of salinity on hydraulic conductivity of compacted bentonite with various dry densities are presented, and analyzed based on the diffusive double layer theory.

## II. Materials and Methods

### 1. Bentonite

The bentonite was a calcium bentonite from Kyungju,

Kyungsangbuk-do, Korea. The chemical composition of bentonite is 53.2 % SiO<sub>2</sub>, 22.1 % Al<sub>2</sub>O<sub>3</sub>, 8.4 % Fe<sub>2</sub>O<sub>3</sub>. It has a cation-exchange capacity of 71 meq/100 g, and Ca<sup>2+</sup> is the predominant exchangeable cation. The bentonite contains montmorillonite (70 %), feldspar (29 %), and small amounts of quartz (~1 %) [8]. The bentonite was passed through a 200 mesh (sieve opening of 0.074 mm) of ASTM standard sieve.

## 2. Experimental

The hydraulic conductivity in the bentonite with a dry density of 1.0 Mg/m<sup>3</sup> to 1.8 Mg/m<sup>3</sup> was measured, which is the range of the compaction density for the engineered barrier in the industrial and radioactive waste disposal facility. The bentonite with an initial water content of 14 wt.% was uniaxially compacted to the desired density in the stainless steel cylindrical cell which has an inside diameter of 50 mm and a height of 25 mm or 10 mm depending on the dry density. The compacted bentonite specimen in the cell was rigidly confined in the chamber by using a restraining ram. The permeant was supplied from the bottom to the top of the chamber at a hydraulic pressure of 300 to 1800 kPa depending on the dry density of bentonite (Fig. 3). Permeants are demineralized water, referred to as freshwater, 0.4 M NaCl solution that is a simulated seawater [9], and 0.04 M NaCl solution that represents the mixture of freshwater and seawater. The penetrated water volumes were measured by weighing. The hydraulic conductivity was determined when equilibrium was reached. In order to

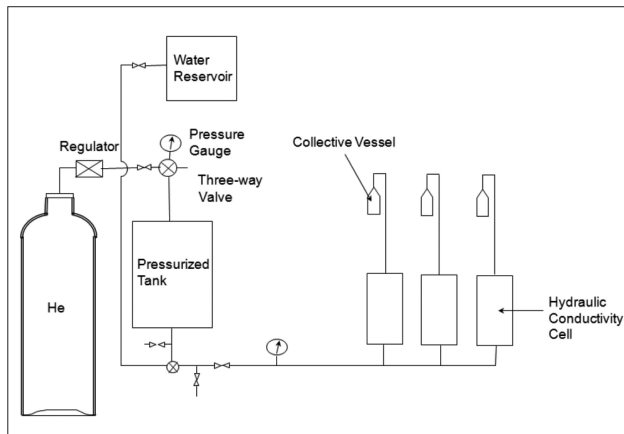


Fig. 3. Schematic diagram of apparatus for measuring hydraulic conductivity of compacted bentonite.

investigate the reversibility of the change of bentonite fabric developed by the intrusion of permeants, two groups of measurements were done for every combination of the dry density of bentonite and the type of permeant. In the first group, saline water is directly introduced into the bentonite specimen, and in the second group, the bentonite specimen was pre-saturated with demineralized water and then saline water was intruded. All measurements were conducted in triplicate.

## II. Results and Discussion

The influence of the NaCl concentration on the hydraulic conductivity of bentonite is shown in Fig. 4 to Fig. 8. The experimental results show that the salinity has an influence on the hydraulic conductivity, and the hydraulic conductivity increases with increasing salinity only when the dry density of

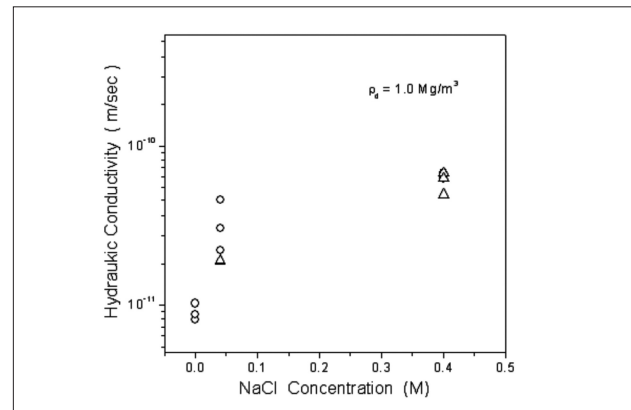


Fig. 4. Hydraulic conductivity of compacted bentonite with dry density of 1.0 Mg/m<sup>3</sup> at different salinity (○: saline water directly introduced, △: demi-water pre-saturated).

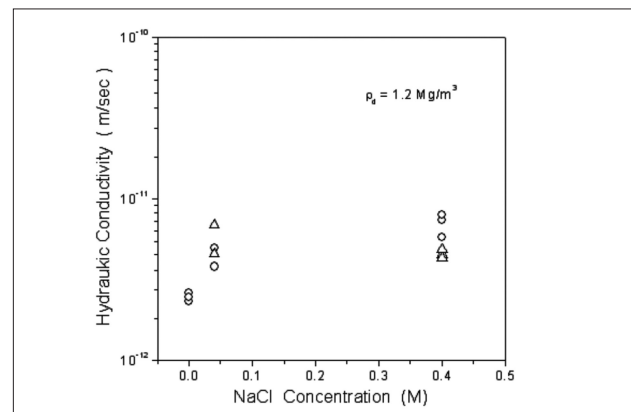
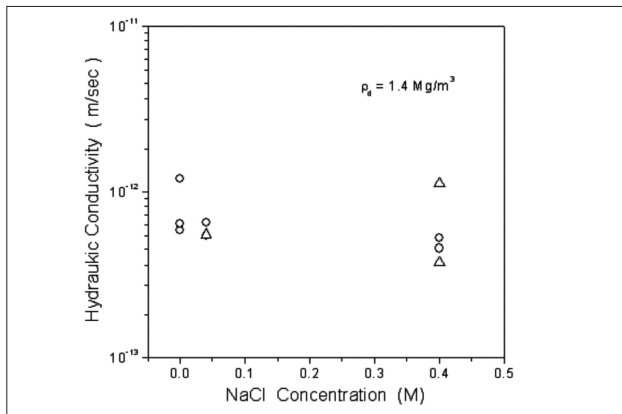
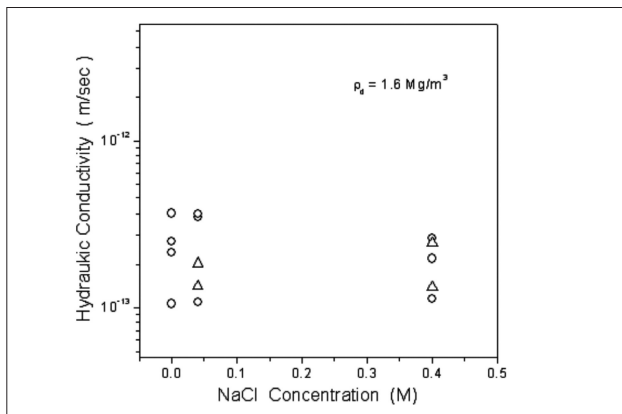


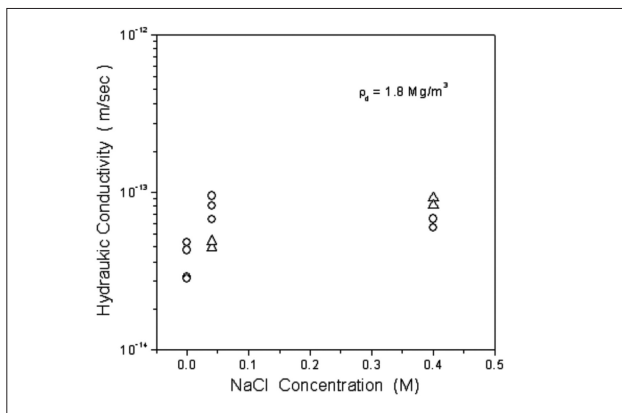
Fig. 5. Hydraulic conductivity of compacted bentonite with dry density of 1.2 Mg/m<sup>3</sup> at different salinity (○: saline water directly introduced, △: demi-water pre-saturated).



**Fig. 6. Hydraulic conductivity of compacted bentonite with dry density of 1.4 Mg/m<sup>3</sup> at different salinity (○: saline water directly introduced, △: demi-water pre-saturated).**



**Fig. 7. Hydraulic conductivity of compacted bentonite with dry density of 1.6 Mg/m<sup>3</sup> at different salinity (○: saline water directly introduced, △: demi-water pre-saturated).**



**Fig. 8. Hydraulic conductivity of compacted bentonite with dry density of 1.8 Mg/m<sup>3</sup> at different salinity (○: saline water directly introduced, △: demi-water pre-saturated).**

bentonite is relatively low. The degree of increase becomes more remarkable at a lower dry density of bentonite. For bentonites with a density of 1.0 Mg/m<sup>3</sup> and 1.2 Mg/m<sup>3</sup>, the hydraulic conductivities of the 0.4 M NaCl solution increase

up to about 7 times and 3 times, respectively higher than that of freshwater. However for the bentonite with a density higher than 1.4 Mg/m<sup>3</sup>, the salinity had an insignificant effect on the hydraulic conductivities, and the hydraulic conductivity is nearly constant within the salinity range of 0 to 0.4 M NaCl. There is also no significant difference between the hydraulic conductivities obtained from the first group measurement that saline water is directly introduced into the bentonite specimen and the second group measurement that the bentonite specimen was pre-saturated with demineralized water and then the saline water was intruded. These results might be rather unexpected because such changes of salinity and the pre-saturation of bentonite with demineralized water are expected to lead to large changes of hydraulic conductivity.

It is known that the hydraulic conductivity of bentonite is affected by the fabric of bentonite [10], and the fabric of bentonite is developed at the time of initial saturation. Bentonite consists principally of montmorillonite that is a swelling material and thus can take up water between their layers. The unit cell in montmorillonite is silica-Gibbsite-silica, and its interlayer bonding is by van der Waals forces. The bonds are weak and easily separated by absorption of water or other polar liquids. Weakness of the interlayer bond is the reason for the high swelling potential. The negative charge associated with montmorillonite particles is caused by an electric imbalance in their molecular configuration. This charge deficiency is satisfied by cations in the pore fluid and polar water molecular that are adsorbed on the montmorillonite surface. In the dry state, a particle of bentonite resembles a closed book composed of many thin crystalline sheets held together. When the freshwater enters into the particles, the fabric of bentonite comes to resemble a pile of crumpled paper. If bentonite is saturated with saline water, the separation distance between the sheets is small and the bentonite particles remain in the form of a closed book [11]. However the experimental results that the pre-saturation of the bentonite specimen with demineralized water has no significant influence on the hydraulic conductivity indicate that the change of the bentonite fabric by intrusion of freshwater or saline water seems to be

reversible. It is intended to explain the effects of salinity change by the relationship between the diffuse double layer and the structure of bentonite. The electric potential is high at the surface where cations are firmly held. However the potential drops sharply with the distance from the montmorillonite surface as the cation diffuse to equalize the concentration. The location where the electrical potential is zero represents an imaginary surface surrounding the montmorillonite particles referred to as the boundary of the diffuse double layer. Montmorillonite contacted with water may cause expansion of the double layer and swelling. The viscosity of the diffusive double layer is higher than that of free water. The water flow is retarded through the diffuse double layers, and it leads to reduced values of hydraulic conductivity.

When equilibrium is established in the double layer, the average local concentration of ions at the distance  $x$  from the surface can be expressed as a function of average electrical potential,  $\Phi$ , at that distance according to Boltzman's theorem;

$$n_- = n_-^* \exp\left(\frac{\nu_- e \Phi}{kT}\right) \dots\dots\dots (1)$$

$$n_+ = n_+^* \exp\left(\frac{-\nu_+ e \Phi}{kT}\right)$$

where  $n_+$  and  $n_-$  are the local concentrations of the positive and negative ions, and  $n_+^*$  and  $n_-^*$  are their concentrations far away from the surface in the equilibrium liquid.  $\nu_+$  and  $\nu_-$  are the valences of the ions.  $e$ ,  $k$  and  $T$  are the elementary charge, the Boltzman constant and the absolute temperature, respectively.

The local electric potential,  $\Phi$  are related by Poisson's equation:

$$\frac{d^2 \Phi}{dx^2} = -\left(\frac{4\pi}{\epsilon}\right) \rho_c \dots\dots\dots (2)$$

where  $\epsilon$  is the dielectric constant,  $\rho_c$  is the local density of charge which is  $\nu_+ n_+ - \nu_- n_-$ . and  $x$  is the distance from the surface.

For simplicity,  $\nu_+ = \nu_-$  is assumed and then  $n_+^* = n_-^* = n$ . From Eq. (1) and (2);

$$\frac{d^2 \Phi}{dx^2} = \left(\frac{8\pi n \nu e}{\epsilon}\right) \sinh\left(\frac{\nu e \Phi}{kT}\right) \dots\dots\dots (4)$$

The following dimensionless groups are introduced for the convenience.

$$y = \frac{\nu e \Phi}{kT} \quad z = \frac{\nu e \Phi_0}{kT} \quad \xi = \kappa x$$

The first integration with the boundary conditions that for  $\xi = \infty$ ,  $dy/d\xi = 0$  and  $y=0$  and the second integration with the boundary conditions that for  $\xi = 0$ ,  $\Phi = \Phi_0$  yield the decay of electric potential,  $\Phi$ , as a function of distance from the particle surface in the diffuse double layer for the clay system as [12]:

$$e^{y/2} = \frac{e^{z/2} + 1 + (e^{z/2} - 1)e^{-\xi}}{e^{z/2} + 1 - (e^{z/2} - 1)e^{-\xi}} \dots\dots\dots (5)$$

$$\kappa^2 = \frac{8\pi n e^2 \nu^2}{\epsilon kT}$$

where  $\epsilon$ ,  $k$ ,  $n$ ,  $e$ ,  $\nu$  and  $T$  are dielectric constant, Boltzman's constant (erg/K), ionic concentration (ions/cm<sup>3</sup>), elementary charge (esu), valence of adsorbed cation and absolute temperature (K), respectively.

For small surface potential ( $\Phi < 25$  mV,  $z < 1$ ) which is the case of bentonite,

$$\frac{d^2 \Phi}{dx^2} = \kappa^2 \Phi \dots\dots\dots (6)$$

$$\Phi = \Phi_0 \exp(-\kappa x) \dots\dots\dots (7)$$

Then the thickness of the diffuse double layer,  $l$  which is the center of gravity of the space charge for the clay system is [12]

$$l = \sqrt{\frac{\epsilon kT}{8\pi n e^2 \nu^2}} \dots\dots\dots (8)$$

When saline water is intruded into bentonite, the cation concentration in pore water is increased, and it causes the diffuse double layer to contract. A reduction in the double layer thickness may result in changes in the net repulsive forces between the montmorillonite sheet which in turn decreases their tendency to separate. When a concentrated brine permeates into clay, shrinkage due to osmotic consolidation occurs. The shrinkage may lead to an alternation of macro and micro structure of the clay, this in

turn causes a change in hydraulic conductivity [13]. Also for the clay permeated with water-soluble organics, the microstructure changes may take place due to the opening of micro pore resulting in an increase in hydraulic conductivity [14,15]. However the experimental results obtained from this study show that change in the hydraulic conductivity is not large and then the microstructural change of bentonite fabric might not occur in the NaCl concentration range of 0 to 0.4 M. Even without particle reorientation, changes of in pore fluid chemistry may alter the thickness of the double diffuse layer. A reduction in double layer thickness may result in a new pathway available for flow and consequently, an increase in hydraulic conductivity. Using Eq. (5), it is possible to calculate the change in electrical potential with increasing distance from the particle surface of Kyungju bentonite for 0.04 M and 0.4 M NaCl solution. The results are shown in

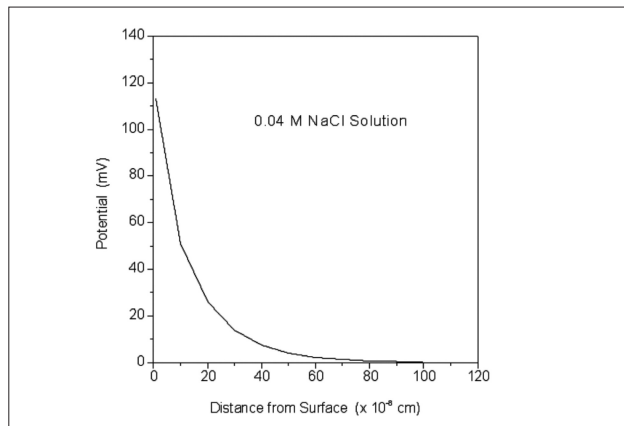


Fig. 9. Variation of electric potential with distance from the surface of the bentonite particle in 0.04 M NaCl solution.

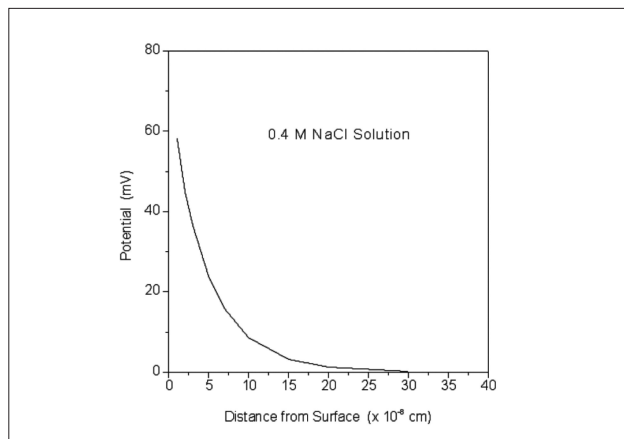


Fig. 10. Variation of electric potential with distance from the surface of the bentonite particle in 0.4 M NaCl solution.

Table 1. Parameters used to calculate the thickness of the diffuse double layer on the surface of bentonite.

parameters	value
Dielectric constant of medium, $\epsilon$	80
Boltzman's constant, $k$ (ergs/ $^{\circ}$ K)	$1.38 \times 10^{-16}$
Elementary charge, $e$ (esu)	$4.80 \times 10^{-10}$
Absolute temperature, $T$ ( $^{\circ}$ K)	293

Fig. 9 and 10. The relationship for the change in the diffuse double layer thickness with changes in cation concentration was developed using Eq. (8). The values of the parameters used in the calculation are listed in Table 1. The thickness of the diffuse double layer is plotted in Fig. 11 for increasing concentration of NaCl. In the case of 0.04 M and 0.4 M NaCl solutions, the thickness of the diffuse double layer are 1.64 nm and 0.48 nm, respectively. Whether or not this contraction of the diffuse double layer will result in the increase of hydraulic conductivity depends on the relationship between the distance between bentonite platelets and the thickness of the diffuse double layer. If the distance between the platelets is small, the diffuse double layers developed from both platelets are still overlapped although they are contracted, and the generation of the effective path for water flow may be insignificant.

The distance between bentonite platelets depends on the dry density of bentonite. The half distance between clay platelets, can be calculated from [16]

$$d = \frac{10^4}{S} \times \left[ \frac{1}{\rho_d} - \frac{1}{\rho_s} \right] \dots\dots\dots (9)$$

where  $\rho_d$  is the clay dry density ( $Mg/m^3$ ),  $\rho_s$  is the specific

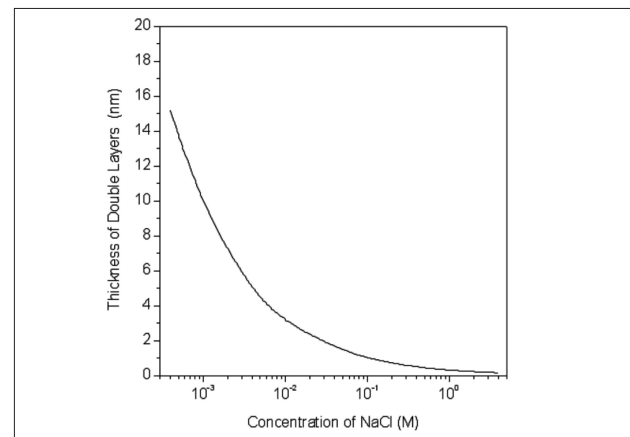
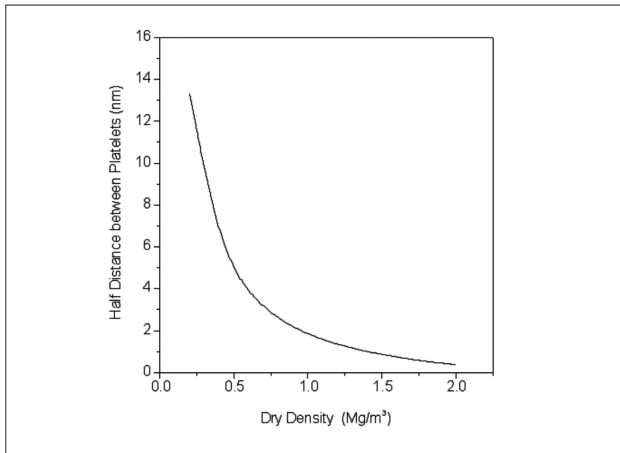


Fig. 11. Thickness of the diffuse double layer versus the concentration of NaCl.



**Fig. 12. Half distance between bentonite platelets as a function of the dry density of bentonite.**

gravity of clay ( $\text{Mg/m}^3$ ), and  $S$  is the specific surface area of the clay ( $\text{m}^2/\text{g}$ ). Using Eq. (9), the half distance between bentonite platelets was calculated and the results are presented in Fig. 12. As shown in the figure, for the dry densities of  $1.0 \text{ Mg/m}^3$  to  $1.8 \text{ Mg/m}^3$ , the half distance between bentonite platelets varies from  $1.81 \text{ nm}$  to  $0.53 \text{ nm}$ . Therefore, if the dry density of bentonite is over  $1.4 \text{ Mg/m}^3$ , the distance between the platelets is so small that the contracted diffuse double layers in  $0.04 \text{ M}$  and  $0.4 \text{ M}$  NaCl solutions are overlapped resulting in nearly constant hydraulic conductivity. When the dry density is lower than  $1.4 \text{ Mg/m}^3$ , the distance between the platelets is relatively large, resulting in the increase of an effective pathway for water flow, and then the hydraulic conductivity increases. The contraction of the diffuse double layer seems to exert a larger influence on water flow as the dry density of bentonite becomes lower.

Eq. (5) and (8) are based on the single flat diffusive double layer theory, and in the compacted bentonite platelets, two flat diffusive double layers are interacted and overlapped. Therefore above discussions cannot describe accurately the real compacted bentonite system. However the application of the interacting flat double layer theory to explain the hydraulic conductivity of the compacted bentonite has not been reported yet, and the difference may be insignificant because the surface charge of the bentonite platelet is small. The application of the single flat diffusive double layer theory can provide an insight and illustration for

the effect of salinity on the hydraulic conductivity of bentonite. The further study is required to apply the interacting flat double layer theory to describe the effects of salinity on the hydraulic conductivity of the compacted bentonite.

### III. Conclusions

The hydraulic conductivities increase with increasing salinity only when the dry density of bentonite is relatively low. The degree of increase becomes more remarkable at a lower dry density of bentonite. For bentonite with the densities of  $1.0 \text{ Mg/m}^3$  and  $1.2 \text{ Mg/m}^3$ , the hydraulic conductivity of the  $0.4 \text{ M}$  NaCl solution increase up to about 7 times and 3 times, respectively higher than those of freshwater. However, for the bentonite with dry densities higher than  $1.4 \text{ Mg/m}^3$ , the salinity has an insignificant effect on the hydraulic conductivities, and the hydraulic conductivity is nearly constant within the salinity range of  $0.04$  to  $0.4 \text{ M}$  NaCl. The pre-saturation of the bentonite specimen with freshwater has no significant influence on the hydraulic conductivity suggesting the reversibility of the change of the bentonite fabric by intrusion of freshwater and saline water. These results will be useful for the optimum design of the high-level waste repository. They are also applicable to the design and reinforcement of low- and intermediate-level radioactive disposal facility and industrial waste landfill site located in a costal area.

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