

## A STUDY ON THE EFFECT OF CLAY PARTICLE ORIENTATION ON DIFFUSION IN COMPACTED BENTONITE

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### ABSTRACT

In this study, the effect of the orientation of clay particles on diffusion in compacted bentonite, which is regarded to be quite important as a candidate buffer material in safety assessment for a geological disposal of high-level radioactive waste, was experimentally discussed by investigating effective diffusion coefficients ( $De$ ) for tritiated water (HTO), which is non-sorptive onto bentonite. The diffusion experiments were carried out for 2 kinds of smectite contents of Na-bentonites, Kunigel-V1® (content of Na-smectite, 46-49wt%) and Kunipia-F® (content of Na-smectite, > 99wt%) at dry densities of 1.0 and 1.5 Mg/m<sup>3</sup> by a through-diffusion method. The through-diffusion experiments were carried out for the same direction as compacted direction of bentonite and perpendicular direction to compacted direction. Scanning electron microscopic (SEM) observations for the cross section of bentonite in the axial and perpendicular directions to compacted direction of bentonite were also carried out at dry densities of 1.0, 1.6, and 2.0 Mg/m<sup>3</sup>. Although  $De$  values for Kunigel-V1® were approximately the same for both diffusion directions to compacted direction over the densities, and no anisotropy in  $De$  was found,  $De$  values in the perpendicular direction to compacted direction for Kunipia-F® were clearly higher than those in the same direction as compacted direction. In the SEM observations, no significant orientation of clay particles was found for Kunigel-V1® over the densities, while the orientation of clay particles was clearly found for Kunipia-F®, and the degree of the orientation of clay particles became significant with an increase in dry density of bentonite. This tendency is in good agreement with that for  $De$  values obtained, indicating that smectite content in bentonite affects the orientation property of clay particles, and that the orientated clay particles affect diffusion pathway.

### INTRODUCTION

In safety assessment of a geological disposal of high-level radioactive waste in Japan, a role as a barrier function of buffer material (bentonite), which composes the multibarrier system of the geological disposal, is quite important, and a lot of studies have been therefore carried out up to the present (e.g. 1). Particularly, since diffusion property of radionuclides in compacted bentonite directly affects the release rate of radionuclides from the bentonite to the geosphere, it is regarded as one of the important parameters in the safety assessment. It is well known from conventional studies that the diffusion of radionuclides in bentonite receives the effects of various pore structural properties or parameters, such as porosity, dry density of bentonite, tortuosity, additives to bentonite (e.g. silica sand), initial bentonite grain size, etc. Since a Na-bentonite, one of the candidate buffer materials, constitutes stacks of smectite flakes, there

is a possibility that the orientation of clay particles takes place in bentonite when the bentonite was compacted, and that the orientated clay particles affect diffusion pathway. The author has preliminarily discussed for the effects of pore structural properties or pore structural factors such as dry density, diffusion direction to compacted direction of bentonite, mixture of silica sand to bentonite, and initial bentonite grain size on diffusion in compacted bentonite in previous studies (2, 3). Moreover, a study on modeling of micropore structure for compacted bentonite has been carried out, and a model of micropore structure considered the orientation of clay particles has been proposed (4). However, neither the orientation property of clay particles in compacted bentonite nor the effect of the orientation property on diffusion has been quantitatively discussed.

In this work, the effect of the orientation of clay particles on diffusion in compacted bentonite is discussed by scanning electron microscopic (SEM) observations and effective diffusion coefficients ( $De$ ) for tritiated water (HTO), which is non-sorptive onto bentonite.

## EXPERIMENTAL

### Diffusion Experiments

The diffusion experiments were carried out by a through-diffusion method (5). Table I shows the experimental conditions for the through-diffusion experiments. The diffusion experiments were carried out for 2 kinds of smectite contents of Na-bentonites, Kunigel-V1® (content of Na-smectite, 46-49wt%) and Kunipia-F® (content of Na-smectite, > 99wt%) at dry densities of 1.0 and 1.5 Mg/m<sup>3</sup>. Both bentonites were obtained from the Kunimine Industries Co. Ltd, Japan. The bentonites were taken from the Tsukinuno mine, Yamagata-ken, Japan. Kunigel-V1® is a crude bentonite, and chalcedony, quartz, plagioclase, calcite, dolomite, analcite, and pyrite are contained as impurities. While Kunipia-F® is a purified bentonite, and no impurity is contained. The detailed mineralogy for these bentonites is described in the literatures of Ito et al. (6, 7).

Figure 1 shows a sectional view of a diffusion cell for a through-diffusion experiment and the enlargement of the sample holder. The diffusion cell consists of a tracer cell, a measurement cell, and a sample holder as shown in Figure 1. The through-diffusion experiments using HTO as a tracer were carried out for the same direction as compacted direction of bentonite and perpendicular direction to compacted direction. Bentonite is filled into the sample holder. At both sides of the sample holder, sintered metal filters sandwich the bentonite to prevent the swelling of the bentonite, and furthermore filter holders sustain the sintered metal filters from outside. The tracer cell is additionally connected with a tracer tank to keep the concentration of HTO in the tracer cell constant, and the solution is circulated by a tubing pump if necessarily. The diffusion experiments were carried out according to the following procedure.

The bentonite was firstly dried at 105°C overnight, and was filled into the sample holder with a cubical space of 15mm. The diffusion experiments in the axial direction to compacted direction were carried out as the compacted bentonite blocks were, while the experiments in the perpendicular direction to compacted direction were performed after the compacted

bentonite blocks were rotated by 90° in angle. The bentonite in the sample holder was then saturated with synthetic porewater for 3 days under atmospheric pressure after being degassed a half hour in a vacuum chamber.

Table I. Experimental Conditions for Diffusion Experiments

Item	Condition
Method	Through-diffusion method
Bentonite	Kunigel-V1® (content of Na-smectite: 46–49wt%) Kunipia-F® (content of Na-smectite: > 99wt%)
Dry density of bentonite	1.0, 1.5 Mg/m <sup>3</sup> (sample size: L15mmxW15mmxH15mm)
Diffusion direction	Axial direction to compacted direction Perpendicular direction to compacted direction
Saturated solution	Synthetic porewater (prepared by NaCl, Na <sub>2</sub> CO <sub>3</sub> , Na <sub>2</sub> SO <sub>4</sub> ) <sup>a</sup>
Tracer solution	Tritiated water (initial concentration: 1.5 kBq/ml)
Temperature	Room temperature (25°C)
Atmosphere	Under air
Diffusing period	12–36 days
Producibility	n=2

<sup>a</sup> See Table II for the chemical composition of the synthetic porewater.

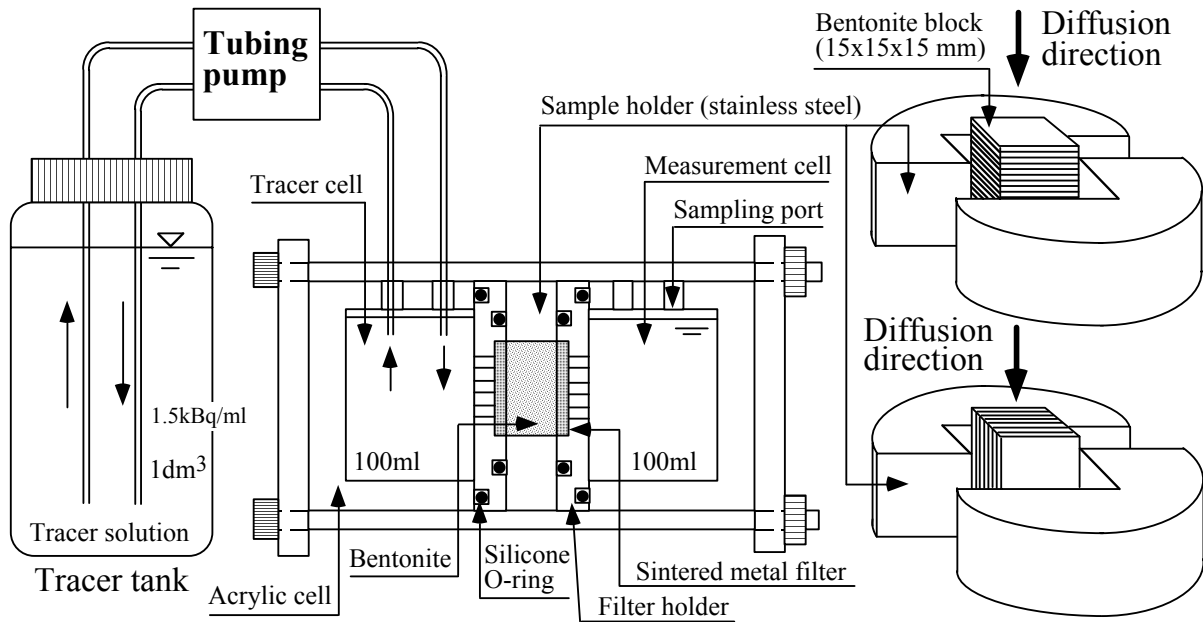


Fig. 1. Sectional view of a diffusion cell and the enlargement of the sample holder.

Table II shows the chemical composition of the synthetic porewater. The synthetic porewater was prepared by NaCl, Na<sub>2</sub>CO<sub>3</sub>, and Na<sub>2</sub>SO<sub>4</sub>. The concentration and the chemical composition of the porewater were determined based on the results of bentonite-water reaction tests for various liquid-solid ratios. After the saturation of the bentonite, the synthetic porewater in the tracer cell was exchanged with that loaded HTO to obtain 1.5 kBq/ml.

Table II. Chemical Composition of Synthetic Porewater

Bentonite	Dry density [Mg/m <sup>3</sup> ]	Concentration [M] <sup>a</sup>				
		Na <sup>+</sup>	Cl <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>	CO <sub>3</sub> <sup>2-</sup>	I. S. <sup>b</sup>
Kunigel-V1	1.0	0.23	0.002	0.024	0.091	0.35
	1.5	0.51	0.0044	0.052	0.20	0.76
Kunipia-F	1.0	0.37	0.021	0.0052	0.17	0.55
	1.5	0.77	0.045	0.011	0.35	1.1

<sup>a</sup> The concentration of each ion was determined by extrapolating the results of bentonite-water reaction tests for various liquid-solid ratios.

<sup>b</sup> I. S.: Ionic strength.

Samples, 0.5ml, were periodically taken from the measurement cell. To minimize the effect of the decrease in the volume of the porewater by sampling, when total volume of the porewater decreased up to 95ml, the porewater in the measurement cell was exchanged with synthetic porewater, in which no tracer is contained.

Through-diffusion experiments of HTO for sintered metal filters were also carried out in order to correct concentration gradients in the filters when  $De$  values are calculated. The concentration of HTO was analyzed for 20 minutes with a liquid scintillation counter. The accumulative quantity of HTO permeated through bentonite specimen from the tracer cell to the measurement cell was obtained with time based on analyzed data. The accumulative quantity is calculated from the following generalized equation.

$$Q_n = C_n \{V - (n-1)v\} + \sum_{i=1}^{n-1} (C_i \cdot v) \quad (n = 1, 2, 3, 4, \dots) \quad (\text{Eq.1})$$

Where  $Q_n$  is the accumulative quantity of the tracer permeated through bentonite specimen up to the  $n$ -th sampling (cpm),  $C_n$  is the analyzed concentration in the  $n$ -th sample (cpm/ml),  $V$  is the solution volume in the measurement cell (ml), and  $v$  is the sampling volume (ml).

At the end of each diffusion experiment, bentonite block in the sample holder was pushed out, and cut with a knife into 1mm pitched slices. Each slice was immediately weighed to accurately obtain the thickness of slice. The slices were immersed in a 2ml HCl solution with a concentration of 0.1M overnight to extract HTO from the slices. Furthermore, a 18ml liquid scintillator was added to individual slices in addition to HCl. The concentrations of HTO were then analyzed, and the concentration profiles in the bentonite were determined based on the analyzed data.

### **Scanning Electron Microscopic (SEM) Observations for the Cross Section of Compacted Bentonite**

Scanning electron microscopic (SEM) observations for the cross section of bentonite in the axial and perpendicular directions to compacted directions were carried out at dry densities of 1.0, 1.6, and 2.0  $\text{Mg/m}^3$  in order to observe the orientation property of clay particles in compacted state. The SEM observations were carried out for both Kunigel-V1® and Kunipia-F® bentonites in order to evaluate the effect of smectite content in bentonite. Figure 2 shows the procedure of the SEM observations. The SEM observations were carried out in the following procedure.

The bentonite was firstly dried at 50°C for 24 hours, and was filled into a stainless column with a cylindrical space of 20mm in diameter and 20mm in thickness. Each bentonite in the column was then saturated with synthetic porewater, of which chemical composition and concentration are the same as that used in diffusion experiments, and was frozen in liquid nitrogen after being pushed out of the column. Each bentonite sample was then dried in a vacuum chamber, and was cut in the axial and perpendicular directions to compacted direction. The SEM observations were carried out for both directions with a magnification of 200 times after being Au-evaporated.

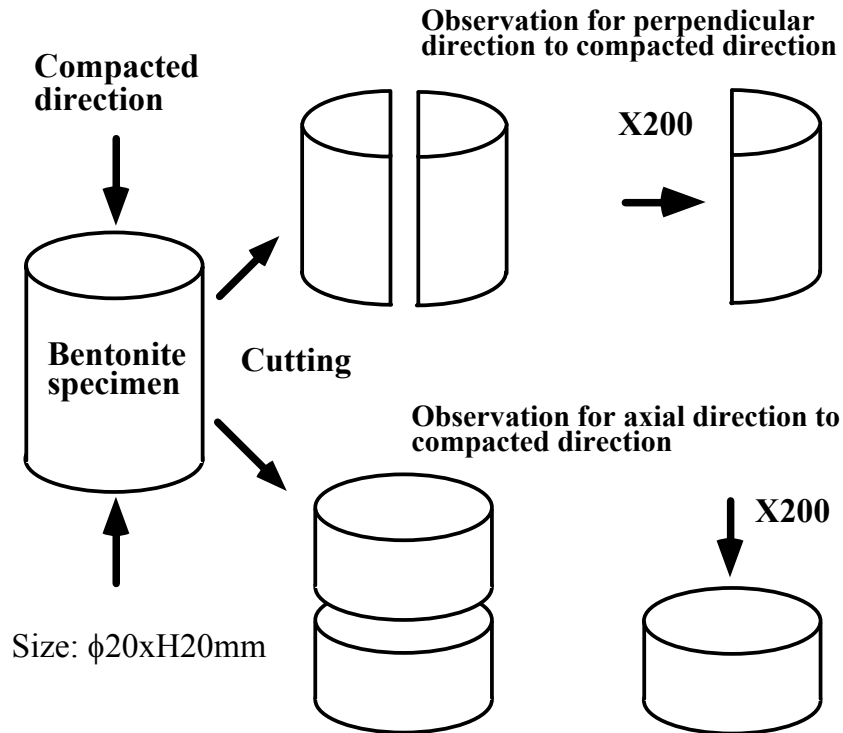


Fig. 2. Procedure of SEM observations for compacted bentonite.

## DIFFUSION THEORY

The calculations of  $De$  values were based on Fickian law (8). The diffusion equation for one-dimensional non-steady state is generally expressed by the following equation (9).

$$\frac{\partial C}{\partial t} = \left( \frac{De}{\alpha} \right) \frac{\partial^2 C}{\partial X^2} \quad (\text{Eq.2})$$

Where  $C$  is the concentration of the tracer in the bentonite ( $\text{cpm}/\text{m}^3$ ),  $t$  is the diffusing time (s),  $De$  is the effective diffusion coefficient ( $\text{m}^2/\text{s}$ ),  $\alpha$  is the rock capacity factor ( $= \phi + \rho_d K_d$ ),  $\phi$  is the porosity of the compacted bentonite,  $\rho_d$  is the dry bulk density of the compacted bentonite ( $\text{Mg}/\text{m}^3$ ),  $K_d$  is the distribution coefficient ( $\text{m}^3/\text{Mg}$ ), and  $X$  is the distance from the tracer source (m).

The  $De/\alpha$  is equal to the apparent diffusion coefficient ( $Da$ ). The accumulative quantity of the tracer permeated through bentonite specimen up to arbitrary time for Eq.(2), based on initial and boundary conditions, is written as follows.

Initial condition

$$C(t, X) = 0, t = 0, 0 \leq X \leq L$$

Boundary condition

$$C(t, X) = C_0 \cdot \alpha, t > 0, x = 0$$

$$C(t, X) = 0, t > 0, X = L$$

$$\frac{Q(t)}{ALCo} = \frac{De}{L} t - \frac{\alpha}{6} - \frac{2\alpha}{\pi^2} \sum_{n=1}^{\infty} \left\{ \frac{(-1)^n}{n^2} \exp\left(-\frac{De n^2 \pi^2 t}{L^2 \alpha}\right) \right\} \quad (\text{Eq.3})$$

Where  $Q(t)$  is the accumulative quantity of the tracer permeated through bentonite specimen (cpm),  $A$  is the cross-sectional area of the bentonite sample ( $m^2$ ),  $L$  is the thickness of the bentonite sample (m), and  $Co$  is the concentration of the tracer in the tracer cell ( $cpm/m^3$ ).

At long time such as steady state, the exponentials in Eq.(3) fall away to zero. Therefore, Eq.(3) is approximately written by the following equation for steady state.

$$\frac{Q(t)}{ALCo} = \frac{De}{L} t - \frac{\alpha}{6} \quad (\text{Eq.4})$$

The  $De$  is calculated from the slope in the change of  $Q(t)$  with time in steady state based on Eq.(4). If surface diffusion does not occur,  $De$  is expressed by the following geometric parameters (9, 10).

$$De = \phi \left( \frac{\delta}{\tau^2} \right) Do = \phi G Do = FF Do \quad (\text{Eq.5})$$

Where  $\delta$  is the constrictivity,  $\tau^2$  is the tortuosity,  $Do$  is the diffusion coefficient in free water ( $m^2/s$ ),  $G$  is the geometric factor (or is also called the tortuosity factor), and  $FF$  is the formation factor.

It is familiar that  $Do$  depends on species in solution. The  $Do$  values for ions are calculated by the Nernst expression as shown below (11).

$$Do = \frac{RT \lambda}{F^2 |Z|} \quad (\text{Eq.6})$$

Where  $R$  is the gas constant (8.314 J/mol/K),  $T$  is the absolute temperature (K),  $\lambda$  is the limiting ionic equivalent conductivity ( $m^2 \cdot S/mol$ ),  $F$  is the Faraday constant (96,493 C/mol), and  $|Z|$  is the absolute value of the ionic charge.

Tritiated water, part of water molecule was exchanged with  $^3H$ , can be regarded as a neutral species. Therefore, although  $Do$  for HTO can not be obtained by electrochemical measurement,  $Do$  directly measured by a diffusion experiment using tracer is reported. In this case, since water diffuses in the water,  $Do$  is called a self-diffusion coefficient, and is reported to be  $2.28E-9 m^2/s$  ( $25^\circ C$ ) for HTO (12).

The concentration gradient of tracer in the filter, which was used to prevent the swelling of bentonite, is also included in  $De$  calculated based on Eq.(4), and must be corrected to calculate true  $De$  in bentonite. This correction is made using the following equation for steady state (13).

$$De = \frac{L}{\left( \frac{L+2L_f}{De_t} \right) - \left( \frac{2L_f}{De_f} \right)} \quad (\text{Eq.7})$$

Where  $De_t$  is the effective diffusion coefficient including the concentration gradient of the tracer in the filter before correction ( $m^2/s$ ),  $De_f$  is the effective diffusion coefficient in the filter ( $m^2/s$ ), and  $L_f$  is the thickness of the filter (m) (1mm in this study).

The detailed derivation process for the correction of  $De$  is described in (14).

## RESULTS AND DISCUSSION

### Effect of Diffusion Direction to Compacted Direction on $De$ and SEM Observations

The concentrations of HTO in the measurement cell as a function of time showed non-linear curves in transient state, and increased in a straight line as a function of time in steady state. The concentration profiles of HTO in bentonite approximately linearly decreased from the tracer cell side to the measurement cell side in all cases. This indicates that the diffusion reached steady state in all cases.

Figure 3 shows  $De$  values of HTO in the axial and perpendicular directions to compacted direction as a function of dry bulk density of bentonite obtained in this study together with  $De$  data of HTO reported to date (15-17). And Table III shows a summary of  $De$  values of HTO obtained in this study together with  $De_f$  values used for correction. All  $De$  data reported up to the present were obtained for axial direction to compacted direction, and are approximately the same as  $De$  values obtained for diffusion in the axial direction to compacted direction in this work. The  $De$  values obtained decreased with increasing dry bulk density of bentonite, showing similar tendency to data reported to date (15-17). The  $De$  values for Kunigel-V1® were approximately the same for both diffusion directions to compacted direction over the densities, and no anisotropy in  $De$  was found, while  $De$  values in the perpendicular direction to compacted direction for Kunipia-F® were clearly higher than those in the same direction as compacted direction, being approximately 4 times greater at a dry density of  $1.5 \text{ Mg/m}^3$ . The  $De$  values for Kunigel-V1® were moreover higher than those for Kunipia-F® on the whole at the same dry density. This is considered to attribute to the difference of smectite partial density (18) caused by the difference of smectite content between both bentonites. The effect of the smectite partial density on diffusion is discussed later. Since the smectite content of Kunipia-F® bentonite is approximately 100wt%, there is a possibility that smectite particles in bentonite orientated in a constant direction by being compacted from one way.



Table III. Summary of  $De$  Values of HTO in Compacted Bentonite Obtained in This Study

Bentonite	Dry density [Mg/m <sup>3</sup> ]	Diffusion direction	Effective diffusion coefficient [m <sup>2</sup> /s]	
			$De_f^a$	$De^b$
Kunigel-V1	1.0	Axial	2.7E-10	3.9E-10
		Perpendicular	2.8E-10	4.1E-10
	1.5	Axial	2.1E-10	2.1E-10
		Perpendicular	2.7E-10	2.6E-10
Kunipia-F	1.0	Axial	3.0E-10	1.2E-10
		Perpendicular	3.1E-10	2.7E-10
	1.5	Axial	3.0E-10	3.3E-11
		Perpendicular	2.5E-10	1.3E-10

<sup>a</sup>  $De_f$ : effective diffusion coefficient in the filter.

<sup>b</sup>  $De$ : effective diffusion coefficient in bentonite.

Axial : diffusion for the same direction as compacted direction.

Perpendicular : diffusion for perpendicular direction to compacted direction.

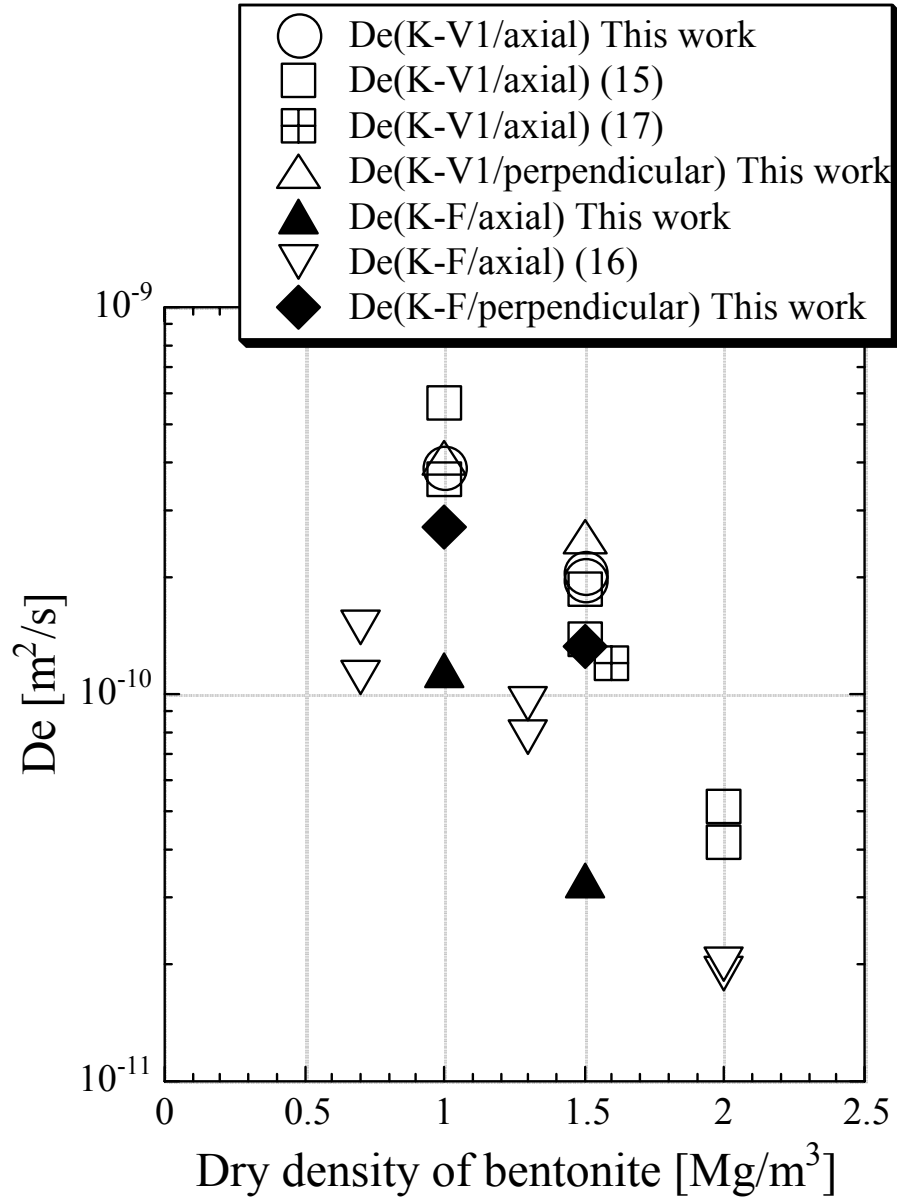


Fig. 3.  $De$  values of HTO for axial and perpendicular directions to compacted direction as a function of dry bulk density of bentonite.

K-V1:Kunigel-V1®, K-F: Kunipia-F®

Axial : diffusion in the axial direction to compacted direction

Perpendicular : diffusion in the perpendicular direction to compacted direction

The difference in micropore structure between Kunigel-V1® and Kunipia-F® was found also in the results of SEM observations. No significant orientation of clay particles was found for Kunigel-V1®, and it was observed that clay particles orientated at random in compacted bentonite. On the other hand, the orientation of clay particles was found for Kunipia-F®, layers of clay particles orientated in the perpendicular direction to compacted direction were observed in the SEM observations, and the degree of the orientation of clay particles became significant with an increase in smectite content in bentonite and dry bulk density of bentonite. Namely, clay particles showed a tendency to orientate in the perpendicular direction to compacted direction of bentonite for Kunipia-F®. This tendency is in good agreement with that for  $De$  values obtained, indicating that smectite content in bentonite affects the orientation property of clay particles, and that the orientated clay particles affect diffusion pathway such as tortuosity.

### Correlation between $De$ Values and Smectite Partial Density

The  $De$  values of HTO for Kunigel-V1®, in which smectite content is about 50wt%, were on the whole higher than those for Kunipia-F®, in which smectite content is approximately 100wt%. Smectite, major clay mineral of bentonite, is considered to compensate spaces between impurities in bentonite. In this case, it is presumed that radionuclides predominantly diffuse in smectite compensating the spaces between the impurities. It is well known also from many literatures reported to date that the diffusivities of radionuclides in smectite depend on the dry bulk density (e.g. 1).

Here the author has considered a density focused on only smectite aggregates, so called the “smectite partial density” (18). This parameter is equivalent to the “montmorillonite density” reported by Kuroda et al. (19). The smectite partial density defined here is more generalized than that proposed by Kuroda et al., and can be applied for all kinds of bentonites. The smectite partial density for a bentonite added additives such as silica sand is generally calculated by the following equation (18, 20).

$$\rho_{dm} = \frac{(1 - fa)fm \cdot \rho_d}{1 - \left\{ \frac{(1 - fa)(1 - fm)}{\frac{1}{n} \sum_{k=1}^n \rho_{imk}} + \frac{fa}{\rho_a} \right\} \rho_d} \quad (\text{Eq.8})$$

Where  $\rho_{dm}$  is the smectite partial density ( $\text{Mg/m}^3$ ),  $\rho_{imk}$  is the solid density of the  $k$ -th impurity ( $\text{Mg/m}^3$ ),  $\rho_a$  is the solid density of the additive added to bentonite ( $\text{Mg/m}^3$ ),  $n$  is the number of the kind of impurity contained in bentonite,  $fa$  is the content of the additive added to bentonite (= additive weight / (additive + bentonite weight)) ( $\text{Mg/Mg}$ ), and  $fm$  is the smectite content in bentonite (= smectite weight / (smectite + impurities weight)) ( $\text{Mg/Mg}$ ).

The detailed derivation process of Eq.8 is described in the literature of (18). With respect to the calculations of  $\rho_{dm}$ , a density of  $2.7 \text{ Mg/m}^3$  was used as the average pure density (solid density) of the impurities. Moreover,  $fm$  for Kunigel-V1® was assumed as 0.5 in the calculations of  $\rho_{dm}$ .

Figure 4 shows a dependency of  $De$  values for HTO on  $\rho_{dm}$ . Data converted from  $Da$  values (1, 15, 16, 21, 22) for HTO to  $De$  values are also simultaneously shown in Figure 4. Since HTO is non-sorptive onto bentonite (1),  $Da$  for HTO can be regarded as diffusion coefficient in porewater ( $Dp$ ), and the relation with  $De$  is given by the following equation.

$$De = \phi \cdot Dp = \left(1 - \frac{\rho_d}{\rho_{th}}\right) Dp \quad (\text{Eq.9})$$

Where  $Dp$  is the diffusion coefficient in porewater ( $m^2/s$ ), and  $\rho_{th}$  is the solid density of bentonite ( $Mg/m^3$ ).

The porosity of compacted bentonite ( $\phi$ ) was calculated assuming  $2.7 Mg/m^3$  (23) as an average solid density of bentonite. Although some variations in  $De$  values are found,  $De$  values for HTO are well correlative with  $\rho_{dm}$ , even though kind of bentonite is different. As is clear also from Figure 4,  $De$  values of HTO for Kunigel-V1® are wholly higher than those for Kunipia-F®. And  $De$  values in the perpendicular direction to compacted direction for Kunipia-F® are higher than the other data. Particularly, significant difference in  $De$  is found at high dry bulk density ( $1.5 Mg/m^3$ ). This is considered to be due to that clay particles orientated in the perpendicular direction to compacted direction for a bentonite, of which smectite content is high such as Kunipia-F®.

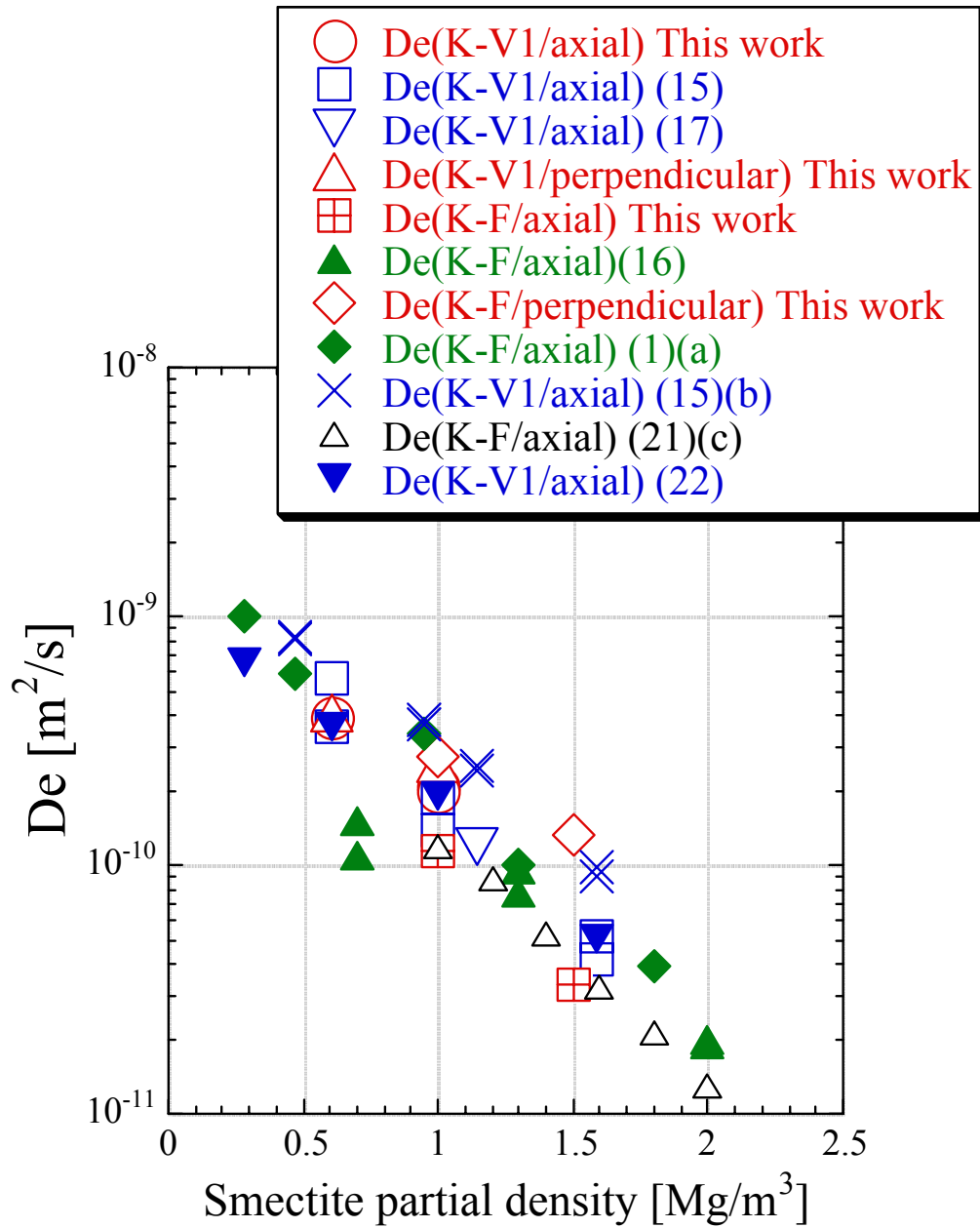


Fig. 4.  $De$  values for HTO as a function of smectite partial density.  
 K-V1 : Kunigel-V1®, K-F: Kunipia-F®  
 Axial : diffusion in the axial direction to compacted direction  
 Perpendicular : diffusion in the perpendicular direction to compacted direction  
 (a), (b), (c) :  $De$  values were calculated from the product of  $Da$  and  $\phi$

## CONCLUSIONS

Through-diffusion experiments using HTO for 2 kinds of smectite contents of Na-bentonites, Kunigel-V1® (content of Na-smectite, 46-49wt%) and Kunipia-F® (content of Na-smectite, > 99wt%) were carried out for the same direction as compacted direction and perpendicular direction to compacted direction at dry densities of 1.0 and 1.5 Mg/m<sup>3</sup>. Scanning electron microscopic (SEM) observations for the cross section of bentonite in the axial and perpendicular directions to compacted direction were carried out at dry densities of 1.0, 1.6, and 2.0 Mg/m<sup>3</sup>. The conclusion is summarized as follows.

- (1) Although *De* values for Kunigel-V1® were approximately the same for both diffusion directions to compacted direction over the densities, and no anisotropy in *De* was found, *De* values in the perpendicular direction to compacted direction for Kunipia-F® were clearly higher than those in the same direction as compacted direction, and the effect of anisotropy in *De* was found.
- (2) The results of SEM observations clearly showed the difference in micropore structure between Kunigel-V1® and Kunipia-F®. No significant orientation of clay particles was found for Kunigel-V1® over the densities, while the orientation of clay particles was found for Kunipia-F®, and the degree of the orientation of clay particles became significant with an increase in dry bulk density of bentonite. This tendency is consistent with that for *De* values obtained, indicating that smectite content in bentonite affects the orientation property of clay particles, and that the orientated clay particles affect diffusion pathway.

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## REFERENCES

1. H. SATO, T. ASHIDA, Y. KOHARA, M. YUI, and N. SASAKI, "Effect of Dry Density on Diffusion of Some Radionuclides in Compacted Sodium Bentonite," *Journal of Nuclear Science and Technology* 29 (9), pp.873-882 (1992).
2. H. SATO, "A Study on Pore Structure of Compacted Bentonite (Kunigel-V1)," JNC TN8400 99-064, Japan Nuclear Cycle Development Institute (1999).
3. H. SATO, "The Effect of Pore Structural Factors on Diffusion in Compacted Sodium Bentonite," *Proc. Int. Symp. Scientific Basis for Nuclear Waste Management XXIV*, Sydney, Australia, August 27-31 2000, Materials Research Society, in press.
4. S. SUZUKI, A. FUJISHIMA, K. UENO, Y. ICHIKAWA, K. KAWAMURA, N. FUJII, M. SHIBATA, H. SATO, and K. KITAYAMA, "Microstructural Modeling of Compacted Sodium-Bentonite and Application of Unified Molecular Dynamics/Homogenization Analysis for Diffusion Process," *Journal of the Clay Science Society of Japan*, 41 (2), pp.43-52 (2001), in Japanese.

5. A. MUURINEN, P. PENTILÄ-HILTUNEN, and J. RANTANEN, "Diffusion Mechanisms of Strontium and Cesium in Compacted Sodium Bentonite," Proc. Int. Symp. Scientific Basis for Nuclear Waste Management X, Boston, Massachusetts, U.S.A., December 1-4 1986, Vol.84, pp.803-811, Materials Research Society (1987).
6. M. ITO, M. OKAMOTO, M. SHIBATA, Y. SASAKI, T. TANBARA, K. SUZUKI, and T. WATANABE, "Mineral Composition Analysis of Bentonite," PNC TN8430 93-003, Power Reactor and Nuclear Fuel Development Corporation (1993), in Japanese.
7. M. ITO, M. OKAMOTO, K. SUZUKI, M. SHIBATA, and Y. SASAKI, "Mineral Composition Analysis of Bentonite," Journal of the Atomic Energy Society of Japan, 36 (11), pp.1055-1058 (1994), in Japanese.
8. J. CRANK, "The Mathematics of Diffusion," 2nd ed., Pergamon Press, Oxford (1975).
9. K. SKAGIUS and I. NERETNIEKS, "Diffusion in Crystalline Rocks of Some Sorbing and Nonsorbing Species," KBS TR82-12, Royal Institute of Technology (1982).
10. H. SATO, T. SHIBUTANI, and M. YUI, "Experimental and Modelling Studies on Diffusion of Cs, Ni and Sm in Granodiorite, Basalt and Mudstone," J. Contaminant Hydrology 26, pp.119-133 (1997).
11. R. A. ROBINSON and R. H. STOKES, "Electrolyte Solutions," 2nd ed., Butterworths, London, p.317 (1959).
12. Chemical Society of Japan, "Chemical Handbook," 4th ed. , Maruzen, Tokyo, p.II-61 (1993), in Japanese.
13. H. SATO, "Effect of Ionic Charge on Effective Diffusion Coefficient in Compacted Sodium Bentonite," Proc. Int. Symp. Scientific Basis for Nuclear Waste Management XXIII, Boston, Massachusetts, U.S.A., November 29-December 2 1999, Vol.608, pp.267-274, Materials Research Society (2000).
14. H. SATO, "Diffusivity Database (DDB) for Major Rocks: Database for the Second Progress Report," JNC TN8400 99-065, Japan Nuclear Cycle Development Institute (1999).
15. H. SATO and T. SHIBUTANI, "Study on Adsorption and Diffusion Mechanism of Nuclides in Buffer Material and Geosphere," PNC Technical Review, No.91, PNC TN 8410 94-284, pp.71-89, Power Reactor and Nuclear Fuel Development Corporation (1994), in Japanese.
16. "Annual Report -FY1989-," PNC TN1410 91-009, pp.51-52, Power Reactor and Nuclear Fuel Development Corporation (1991), in Japanese.
17. H. KATO and T. YATO, "Estimation of the Effective Diffusivity in Sand/Bentonite Mixture," 1997 Fall Meeting of the Atomic Energy Society of Japan, October 14-17 1997, I39, Atomic Energy Society of Japan (1997), in Japanese.
18. H. SATO and S. MIYAMOTO, "A Study on Diffusion and Migration of Lead in Compacted Bentonite," JNC TN8400 2001-018 , Japan Nuclear Cycle Development Institute (2001).
19. Y. KURODA, K. IDEMITSU, H. FURUYA, Y. INAGAKI, and T. ARIMA, "Diffusion of Technetium in Compacted Bentonites in the Reducing Condition with Corrosion Products of Iron," Proc. Int. Symp. Scientific Basis for Nuclear Waste Management XX, Boston, Massachusetts, U.S.A., December 2-6 1996, Vol.465, pp.909-916, Materials Research Society (1997).

20. H. SATO and S. MIYAMOTO, "The Effect of Silica Sand Content and Temperature on Diffusion of Selenium in Compacted Bentonite Under Reducing Conditions," Proc. Int. Symp. Scientific Basis for Criticality Safety, Separation Process and Waste Disposal, Tokai-mura, Ibaraki, Japan, October 31-November 2 2001, Japan Atomic Energy Research Institute, in press.
21. Y. TORIKAI, Doctoral Thesis of Hokkaido University (1996), in Japanese.
22. H. KATO, M. MUROI, N. YAMADA, H. ISHIDA, and H. SATO, "Estimation of Effective Diffusivity in Compacted Bentonite," Proc. Int. Symp. Scientific Basis for Nuclear Waste Management XVIII, Kyoto, Japan, October 23-27 1994, Vol.353, pp.277-284, Materials Research Society (1995).
23. H. SUZUKI, M. SHIBATA, J. YAMAGATA, I. HIROSE, and K. TERAOKA, PNC TN8410 92-057, Power Reactor and Nuclear Fuel Corporation (1992), in Japanese.