

## Diffusion Characteristics of Iodide in a Domestic Bentonite of Korea

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### 국산벤토나이트에서의 요오드이온의 확산특성

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#### Abstract

The transport of radionuclides in a compacted bentonite is dominated by diffusion. Through diffusion tests for iodide were performed to investigate the diffusion characteristics of anionic radionuclides in a domestic bentonite. The bentonite used was sampled from the southeastern area of Korea and the solution was synthetic groundwater spiked with a tracer of I-125 (as Na<sup>125</sup>I). The dry densities of compacted bentonite were 1.2, 1.4, and 1.7 Mg/m<sup>3</sup>. The apparent diffusion coefficients and the effective diffusion coefficients of the iodide decrease with increasing dry density. The values were from  $3.80$  to  $7.12 \times 10^{-11}$  m<sup>2</sup>/s for the apparent diffusion coefficients and from  $1.25$  to  $7.97 \times 10^{-12}$  m<sup>2</sup>/s for the effective diffusion coefficients, respectively. The experimental results also showed that the apparent diffusion coefficients depended on the pore structure of compacted bentonite and the effective diffusion coefficients were attributed to the pore structure and the effective porosity that represents the available pathway for the diffusional transport of iodide. The results obtained will be used as basic data for the safety assessment of a repository.

#### 요 약

압축 벤토나이트에서 핵종이동은 확산에 의해 지배된다. 국산 벤토나이트에 대한 음이온핵종의 확산 특성 규명을 위해서 요오드이온을 대상으로 관통확산실험을 수행하였다. 대상매질은 동해안 지역의 벤토나이트를 사용하였으며, 실험용액은 합성 모의지하수에 I-125를 첨가하여 사용하였다. 압축 벤토나이트의 충전밀도는 1.2, 1.4, 1.7 Mg/m<sup>3</sup> 등 세가지로 하였다. 요오드이온의 겔보기확산계수와 유효확산계수는 충전밀도가 증가함에 따라 감소하였고, 이들 확산계수의 값은 각각  $3.80 - 7.12 \times 10^{-11}$  m<sup>2</sup>/s와  $1.25 - 7.97 \times 10^{-12}$  m<sup>2</sup>/s의 범위에 있었다. 실험결과, 겔보기확산계수는 벤토나이트의 공극구조에 의존하였으며, 유효확산계수는 공극구조는 물론 핵종확산이동에 이용된 유효공극율에 의해 좌우되었다. 본 연구에서 얻어진 결과는 처분장안전성평가의 기초자료로 활용될 것이다.

## 1. Introduction

Bentonite, a natural clay is widely favored as a backfill material of repository to retard the release of radionuclides from the radwaste to the surrounding environment, because it has very low hydraulic conductivity and relatively high sorption capacity.

When the bentonitic clay is used for the backfill, the hydraulic conductivity is generally below  $10^{-11}$  m/s and the principal transport mechanism of radionuclides is known to be diffusion [1, 2]. Understanding the diffusion characteristics of radionuclides in the compacted bentonite is, therefore, of essence in the assessment of radionuclide release through the backfill of a repository.

The diffusion of radionuclides in compacted bentonite has been studied by many investigators [3-9, 16-18]. According to their studies, different radionuclides seemed to follow different diffusion characteristics. Most anionic radionuclides migrated quickly without sorption onto the bentonite, and they had different diffusion mechanisms from cationic radionuclides. Little has been reported about the information on the diffusion of the anionic radionuclides through a domestic bentonite available in Korea.

In this paper, iodide is selected to study the diffusion behavior of anionic radionuclides in the compacted bentonite. The diffusion coefficients of the iodide in a domestic bentonite are measured for various dry densities by a through-diffusion test, and the diffusion characteristics are also investigated to enhance the understanding of the migration phenomena in a compacted bentonite.

## 2. Theoretical Background

Diffusion is the process by which a matter is transported from one part of a system to another by random molecular motions. The mathematical description of diffusion is based on the Fick's I and II laws of diffusion, which are expressed as follows:

$$\tilde{F} = -D_o \nabla C \quad (1)$$

$$\frac{\partial C}{\partial t} = -\nabla \cdot \tilde{F} = D_o \nabla^2 C \quad (2)$$

where  $\tilde{F}$  is the diffusive flux,  $C$  the concentration,  $D_o$  the diffusion coefficient, and  $t$  the time. The above equations are applied to a homogeneous and isotropic medium such as free-water.

However, the Fick's I and II laws of diffusion should be modified for a porous medium, because there are several diffusion mechanisms and interactions between solid and liquid phases. If the transport of a matter through a porous medium is controlled by diffusion, the equations (1) and (2) are modified as

$$\tilde{F}_p = -D_a \nabla \bar{C} \quad (3)$$

$$\frac{\partial \bar{C}}{\partial t} = -\nabla \cdot \tilde{F}_p = D_a \nabla^2 \bar{C} \quad (4)$$

where  $\tilde{F}_p$  is the diffusive flux based on the cross sectional area of porous medium,  $\bar{C}$  the average concentration of a diffusant over the solid and the liquid phases,  $D_a$  the apparent diffusion coefficient [10].

In the equations (3) and (4),  $D_a$  is not a molecular diffusion coefficient in free water but denotes an apparent diffusion coefficient in porous medium. The apparent diffusion coefficient is generally a function of many factors such as molecular diffusion, pore geometry, surface diffusion and physicochemical processes including adsorption, ion-exchange, and precipitation.

The apparent diffusion coefficient may therefore be defined in several forms depending upon the factors taken into account for the diffusion through the porous medium. For sorbing radionuclides it has been expressed in two forms in the referenced literature [1, 11]. When the radionuclides sorbed on the particle surfaces of a medium migrate along the surface,  $D_a$  is defined as follows:

$$D_a = \frac{D_p \varepsilon + D_s \rho_b K_d}{\varepsilon + \rho_b K_d} \quad (5)$$

where  $D_p$  is the pore diffusion coefficient,  $\varepsilon$  the po-

rosity of the porous medium,  $\rho_b$  the bulk density,  $D_s$  the surface diffusion coefficient, and  $K_d$  the sorption distribution coefficient. The so-called effective diffusion coefficient  $D_e$  is expressed:

$$D_e = D_a(\epsilon + \rho_b K_d) = D_p \epsilon + D_s \rho_b K_d \quad (6)$$

If there is no surface diffusion ( $D_s = 0$ ), the equations (5) and (6) are respectively given as

$$D_a = \frac{D_p \epsilon}{\epsilon + \rho_b K_d} \quad (7)$$

$$D_e = D_a(\epsilon + \rho_b K_d) = D_p \epsilon \quad (8)$$

When there are neither sorption nor surface diffusion ( $K_d = 0$ ,  $D_s = 0$ ) like anion, on the other hand, the equations (5) and (6) have very simple forms:

$$D_a = D_p = D_o f \quad (9)$$

$$D_e = D_a \epsilon = D_p \epsilon = D_o f \epsilon \quad (10)$$

where  $D_o$  is the molecular diffusion coefficient in free water and  $f$  is the geometrical factor of the porous medium.

For the measurement of the diffusion coefficients in compacted bentonite, several experimental techniques have been used: through-diffusion, in-diffusion, back-to-back diffusion, and reservoir-depletion method [1, 11]. This study used the through-diffusion method to determine the diffusion coefficients of iodide in a domestic bentonite. A compacted bentonite saturated with water is sandwiched between two reservoirs. Initially, one of the reservoirs contains tracer, whereas the other bentonite plug is free of tracer.

The tracer diffuses from the high concentration reservoir through the compacted bentonite into the other reservoir where the concentration is monitored. The concentration gradient is maintained from the reservoir initially containing tracer to the other reservoir.

Figure 1 schematically represents the configuration of radionuclide transport in a through-diffusion method. If the compacted layers of bentonite are homogeneous and isotropic and the transport of

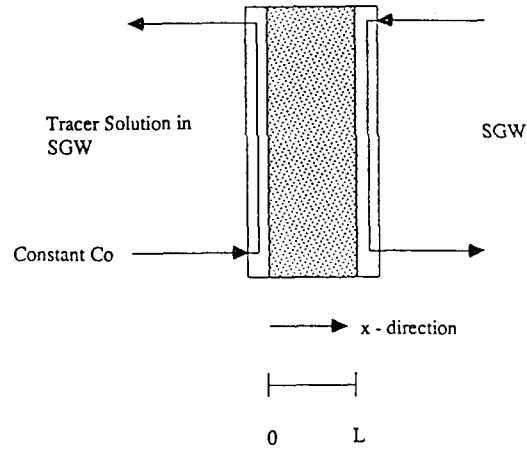


Fig. 1. Schematic Representation of Radionuclide Migration in the Through-Diffusion Method

radionuclide occurs only in the  $x$ -direction, the equation (4) is expressed in a simplified form.

$$\frac{\partial \bar{C}}{\partial t} = D_a \frac{\partial^2 \bar{C}}{\partial x^2} \quad (11)$$

The following initial and boundary conditions are given to describe the experimental constraints of the through-diffusion method

$$\bar{C}(x, 0) = 0 \quad \text{for } 0 < x < L \quad (12)$$

$$\bar{C}(0, t) = \bar{C}_o \quad \text{for } t > 0 \quad (13)$$

$$\bar{C}(L, t) \ll \bar{C}_o \quad \text{for } t > 0 \quad (14)$$

where  $\bar{C}_o$  is the initial radionuclide concentration at the interface between the tracer-containing reservoir and the bentonite plug, and  $L$  the overall length of the bentonite plug.

The solution of the equation (11) subject to the initial and boundary conditions from equations (12) to (14) was given by Helfferich [5, 12] as

$$\bar{C}(x, t) = \bar{C}_o \left(1 - \frac{x}{L}\right) - \sum_{n=1}^{\infty} 2 \frac{\bar{C}_o}{n\pi} \exp\left(-\left(\frac{n\pi}{L}\right)^2 D_a t\right) \sin\left(\frac{n\pi}{L} x\right) \quad (15)$$

The total amount,  $Q$  of radionuclide accumulated in the solution at the far side of the bentonite plug is obtained by integrating the flux at  $x = L$  with time:

$$Q/\bar{C}_0 A = \frac{D_a t}{L} - \frac{L}{6} - \frac{2L}{\pi^2} \sum_{n=1}^{\infty} \frac{(-1)^n}{n^2} \exp\left(-\frac{n^2 \pi^2 D_a t}{L^2}\right) \quad (16)$$

Defining  $\bar{C}_0 = (\varepsilon + \rho_b K_d) C_0 = \alpha C_0$  where  $C_0$  represents the initial concentration of the spiked reservoir, the equation (16) can be expressed as

$$Q/C_0 A = \frac{\alpha D_a t}{L} - \frac{\alpha L}{6} - \frac{2\alpha L}{\pi^2} \sum_{n=1}^{\infty} \frac{(-1)^n}{n^2} \exp\left(-\frac{n^2 \pi^2 D_a t}{L^2}\right) \quad (17)$$

where  $\alpha$  is referred to as a capacity factor, which is defined as  $\alpha = \varepsilon + \rho_b K_d$  for sorbing radionuclide and as  $\alpha = \varepsilon$  for nonsorbing radionuclide [11].

A typical breakthrough curve of the eqn. (17) is shown in Figure 2. Initially, there is a transient period during which the concentration builds up at the plug, and then there will be a stage of steady-state diffusion during which the amount of radionuclide transported through the plug is given by the asymptotic solution

$$Q/C_0 A = \frac{\alpha D_a t}{L} - \frac{\alpha L}{6} \quad (18)$$

as the diffusion time approaches to an infinite. From the equation (6), the relationship of  $D_a = D_e / \alpha$  is obtained, where  $\alpha$  is the same as in the equation (17). Thus, the equation (18) becomes

$$Q/C_0 A = \frac{D_e t}{L} - \frac{\alpha L}{6} \quad (19)$$

From the equation (18), the apparent diffusion coefficient  $D_a$  can be obtained by using an intercept  $t_{lag}$  (time-lag) on the time-axis, i.e., by setting the left-hand side of the equation (18) equal zero:

$$D_a = \frac{L^2}{6 t_{lag}} \quad (20)$$

The effective diffusion coefficient is obtained from the slope,  $S$  of steady-state diffusion in the equation (19) as follows:

$$S = \frac{D_e}{L} \quad \text{or} \quad D_e = S L \quad (21)$$

### 3. Experiment

#### 3.1. Material

The domestic bentonite was sampled from the southeastern area of Korea. The bentonite was air-dried and passed through a 300 mesh of ASTM standard sieves. As shown in the result of X-ray diffraction analysis (Figure 3), the bentonite contains mainly montmorillonite and small amounts of feldspar, quartz, and zeolite. The chemical composition of the bentonite is summarized in Table 1: about 53%  $\text{SiO}_2$ , 22%  $\text{Al}_2\text{O}_3$ , 8%  $\text{Fe}_2\text{O}_3$ , and minor amounts of  $\text{FeO}$ ,  $\text{CaO}$ ,  $\text{MgO}$ ,  $\text{K}_2\text{O}$ ,  $\text{Na}_2\text{O}$ , and  $\text{MnO}$ . The weight percent of calcium was about 2 times higher than that of sodium, and the cation exchange capacity was 70.9 meq/100g.

#### 3.2. Radionuclides and Solution

In aqueous solution, iodine exists in several forms depending on the pH and Eh (redox potential) of

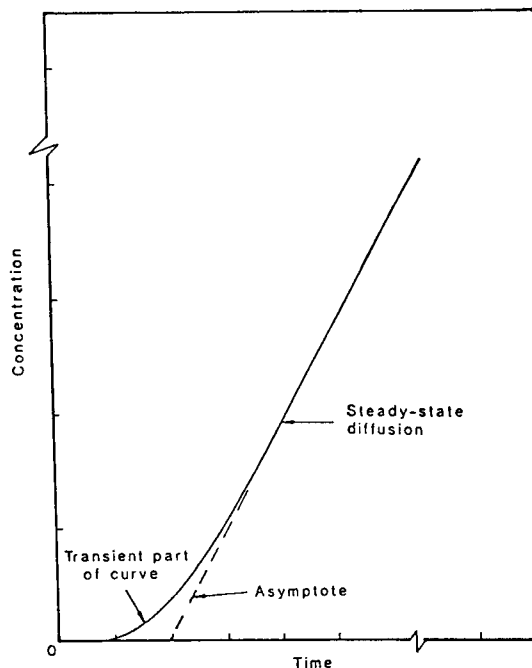


Fig. 2. Typical Breakthrough Curve of Radionuclide in the Through-Diffusion Method

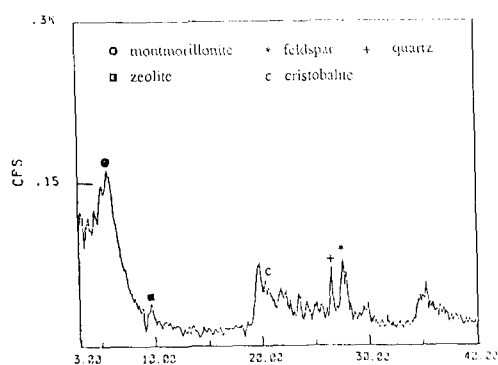


Fig. 3. X-Ray Diffraction Pattern of the Domestic Bentonite Used in the Diffusion Test

Table 1. Chemical Composition of a Domestic Bentonite from the Southeastern Area of Korea

COMPONENT	CONTENT (WT. %)
SiO <sub>2</sub>	53.20
Al <sub>2</sub> O <sub>3</sub>	22.05
Fe <sub>2</sub> O <sub>3</sub>	8.37
FeO	0.32
CaO	2.63
MgO	1.98
K <sub>2</sub> O	0.96
Na <sub>2</sub> O	1.36
MnO	0.11
IG - Loss	

the solution. Lieser [13] reported that three species of  $I^-$ ,  $IO_3^-$ , and  $I_2$  had to be considered at a low concentration if organic compounds that may react with iodine are excluded. The iodide was, in particular, stable over a wide range of pH and Eh in ground water, so that it was selected as an anion of interest in this study. The  $^{125}I$  in the form of  $Na^{125}I$  was chosen as a tracer. A synthetic groundwater was used to saturate the bentonite. The elemental composition and pH of the synthetic groundwater are shown in Table 2. Tracer solutions were prepared by adding a small aliquot of I-125 with chemical form

Table 2. Elemental Composition of Synthetic Groundwater Use in the Diffusion Tests[19]

COMPONENT	CONCENTRATION(ppm)
Na	8.3
K	3.5
Mg	3.9
Ca	13.0
HCO <sub>3</sub>	ND
Cl	5.0
SO <sub>4</sub>	8.6
NO <sub>3</sub>	0.62
F	0.19

\* pH = 7.0 + / - 0.5

of NaI to synthetic groundwater. The initial activity of I-125 was 0.1 Ci/m<sup>3</sup>.

### 3.3. Through-Diffusion Test

The test was performed to investigate the diffusion behavior of iodide in a domestic bentonite. Figure 4 shows an apparatus used in the through-diffusion test. As shown in the figure, it consists of three parts, a source reservoir, a collection reservoir, and a cylindrical diffusion cell made of stainless steel with  $4 \times 10^{-2}$  m of inner diameter and  $1.2 \times 10^{-2}$  m of height. The experimental procedure is as follows. The air-dried bentonite was compacted in the diffusion cells to the target dry densities of 1.2, 1.4, and 1.7 Mg/m<sup>3</sup> by a hydraulic press. The diffusion cells were placed in a housing and the bentonite was allowed to be saturated with SGW (Synthetic Ground Water) for 6 weeks. Nowak[14] reported that this period was sufficient to saturate the bentonite. After the saturation period, a solution of SGW spiked with I-125 was passed over one end of the bentonite plug and an unspiked SGW was circulated through the other end. The flow rate of the unspiked solution was such that the activity of I-125 would be very close to zero at the outflow end of the plug. The col-

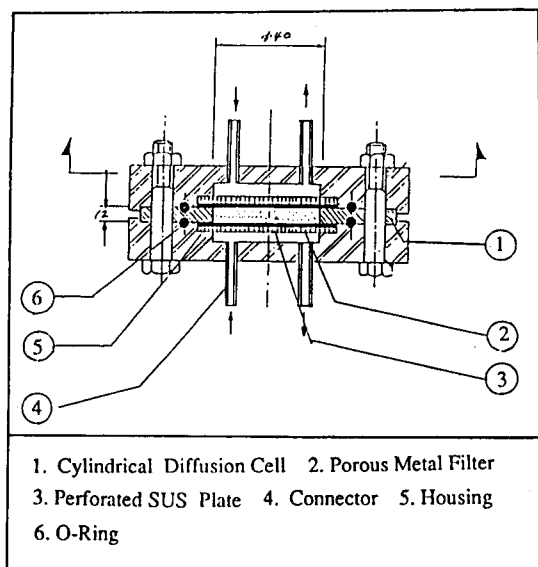


Fig. 4. Experimental Apparatus for the Through-Diffusion Test

lection reservoir was monitored every two or three days by a liquid scintillation counter. The activity in the source reservoir was also closely monitored. When it approached about 90% of the starting activity, I-125 was added to bring its activity up to that of the original solution. The experiment was ended several days after the steady-state was reached. During the experiment the temperature was maintained at about 20°C.

#### 4. Results and Discussion

Sorption test was carried out by a batch method [17] to investigate the interaction of iodide with bentonite. The ratio of bentonite to solution was 0.1 Mg/10m<sup>3</sup> and the concentration ranged from 10<sup>-12</sup> to 10<sup>-4</sup> M. The contacting time for the equilibrium between liquid and solid phases was four weeks. The results showed that there was no sorption of iodide on the bentonite and, if any, it was expected to be negligible. One possible explanation for no sorption behavior is an anion exclusion. The iodides around

the bentonite particle are excluded by the negative surface charge of the bentonite and hence they do not adsorb onto the bentonite. The following phenomenon gave a proof of the anion-exclusion. When the bentonite was separated from the solution by centrifuging, the bentonite particles were pressed close to one another at the bottom of the centrifuge tube, and in consequence the concentration of iodide in the solution separated by centrifuging at the end of the sorption test increased by 2.0–11.0% of the initial concentration due to the simultaneous interference of anion exclusion and very low sorption. Similar phenomenon was also observed in Muurinen's sorption test [15] with sodium bentonite–chloride system.

The diffusion of iodide in the compacted bentonite with the dry densities of 1.2, 1.4, and 1.7 Mg/m<sup>3</sup> was shown in Figure 5. In the figure the breakthrough curves became asymptotic lines after a relatively long period of diffusion. The apparent diffusion coefficients and the effective diffusion coefficients were calculated by the equations (20) and (21) from the time-lags and slopes of the asymptotic lines.

Table 3 showed that the apparent diffusion coefficients of iodide in the compacted bentonite were in the range of 7.12–3.80 × 10<sup>-11</sup> m<sup>2</sup>/s, which were two order of magnitude lower than those in

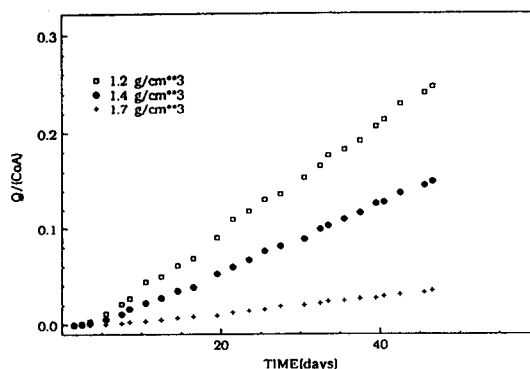


Fig. 5. Breakthrough Curves of Iodide in the Compacted Bentonite with the Dry Densities of 1.2, 1.4, 1.7 Mg/m<sup>3</sup>

**Table 3. Diffusion Properties of Iodide in the Compacted Bentonite for Various Dry Densities**

$\rho_b$	$\epsilon$	Da	De	f *	$\epsilon_e$	$(\epsilon_e/\epsilon) \times 100$
(g/cm <sup>3</sup> )	(-)	( $\times 10^{-11}$ m <sup>2</sup> /sec)	( $\times 10^{-12}$ m <sup>2</sup> /sec)	(-)	(-)	(%)
1.2	0.56	7.12	7.97	0.039	0.11	20
1.4	0.48	5.80	4.99	0.032	0.09	19
1.7	0.37	3.80	1.25	0.021	0.03	8

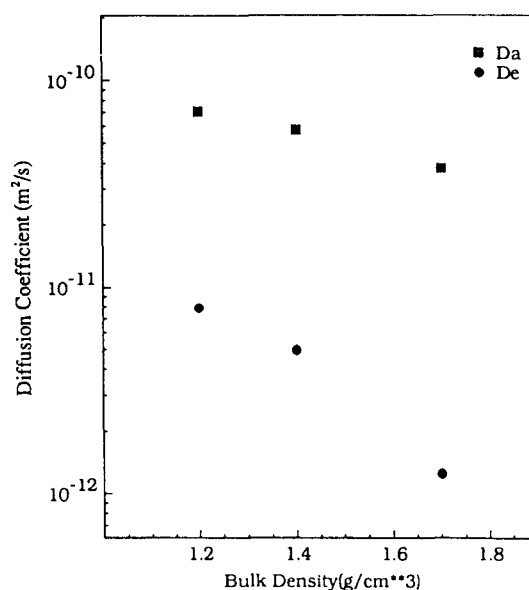
\*  $D_0 = 1.79 \times 10^{-9}$  m<sup>2</sup>/sec for iodide

pure bulk solution. The apparent diffusion coefficients decreased with increasing the dry density. The decrease in  $D_a$  was attributed to a decrease in the geometrical factor  $f$  ( $f = \delta/\tau^2$ ), where  $\tau$  and  $\delta$  were tortuosity and constrictivity, respectively. It suggests that the diffusion of iodide depends upon the pore structure of the compacted bentonite. When the bentonite is compacted, the tortuosity and the constrictivity of the diffusion path around bentonite particles seem to increase and consequently give the decrease in the geometrical factor and the apparent diffusion coefficient.

In the table 3, the effective diffusion coefficients of iodide in the compacted bentonites ranged from  $1.23 \times 10^{-12}$  m<sup>2</sup>/s at  $\rho_b = 1.7$  Mg/m<sup>3</sup> to  $7.97 \times 10^{-12}$  m<sup>2</sup>/s at  $\rho_b = 1.2$  Mg/m<sup>3</sup>. The values were one order of magnitude lower than those of the apparent diffusion coefficients. The effective diffusion coefficients also decreased with increasing the dry density of the compacted bentonite. An increase in the dry density gave an increase in the anionic exclusion volume, and it resulted in a decrease in  $\epsilon_e$  which represented the effective porosity of the compacted bentonite available for the diffusion of iodide. The effective porosity was shown to be approximately 8–20% of the total porosity of the compacted bentonite with the dry densities of 1.2–1.7 Mg/m<sup>3</sup>. The decrease in  $\epsilon_e$  can be explained in terms of anion exclusion phenomenon that the hydrated iodides are excluded from the bentonite particle surfaces. An increase in

the dry density gives an increase in the anionic exclusion volume, which results in a decrease in the effective porosity in the compacted bentonite.

Figure 6 showed that the  $D_e$  values were more sensitive to the dry density than the  $D_a$  values. It was attributed to a decrease in the effective porosity of the compacted bentonite. From this, we can speculate that at the high dry density most of the smaller pores in the compacted bentonite are not available



**Fig. 6. The Apparent and Effective Diffusion Coefficients of Iodide in the Compacted Bentonite for Different Dry Densities**

for the diffusion of the hydrated iodide due to the anion exclusion, and thus the iodide moves only through the larger pores which may be relatively small portion.

As discussed above, we can recognize that the effective porosity is a critical parameter in the diffusion of iodide through compacted bentonite. Figure 7 gave a comparison of the diffusive fluxes for the total porosity and the effective porosity in the bentonite with a typical dry density of  $1.4 \text{ Mg/m}^3$  using the equation (17). As shown in the figure, there was a large difference between the diffusive fluxes calculated for the total porosity and the effective porosity. It indicated that the effective porosity should be used to predict the diffusive fluxes of iodide through the compacted bentonite backfill of a repository.

### 5. Conclusion

Sorption tests and through-diffusion tests were carried out for the iodide to investigate the diffusion behavior of anionic radionuclide in a domestic compacted bentonite. It was revealed that there was no sorption of iodide onto the bentonite in the batch

method. When the dry densities of the compacted bentonite ranged from  $1.2$  to  $1.7 \text{ Mg/m}^3$ , the apparent diffusion coefficients and the effective diffusion coefficients were in  $3.80 - 7.12 \times 10^{-11} \text{ m}^2/\text{s}$  and  $1.25 - 7.97 \times 10^{-12} \text{ m}^2/\text{s}$ , respectively. Both the diffusion coefficients decreased with increasing the dry density of the compacted bentonite. The extent of compaction had more influence on the effective diffusion coefficient than on the apparent diffusion coefficient. The apparent diffusion coefficients were dependent on the pore structure of compacted bentonite. The decrease in the effective diffusion coefficients were attributed to a decrease in the effective porosity as well as the effect of pore structure. The effective porosity seemed to be controlled by anion exclusion. The experimental results suggested that the effect of dry density on the effective porosity should be taken into account in predicting the diffusive fluxes of anion such as iodide in the bentonitic barrier of a repository.

### Acknowledgment

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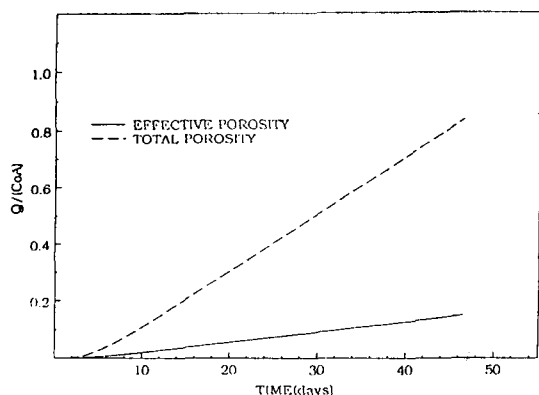


Fig. 7. Diffusive Fluxes of Iodide for the Effective and Total Porosities in the Compacted Bentonite with a Typical Dry Density of  $1.4 \text{ Mg/m}^3$  (Predicted by the Eqn.(17))



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