

## Radionuclide Diffusion in Compacted Domestic Bentonite

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### ABSTRACT

The diffusion of Sr-85, Cs-137, Co-60 and Am-241 in compacted domestic bentonite was studied, using a diffusion cell unit in which diffusion took place axially from the center of cylindrical bentonite sample body. The effects of compaction density and heat-treated bentonite on diffusion were analysed. And the diffusion mechanism of radionuclide was also analysed by evaluating the measured diffusivity of anion Cl-36.

The apparent diffusivities obtained for Sr-85, Cs-137, Co-60 and Am-241 were  $1.07 \times 10^{-11}$ ,  $6.705 \times 10^{-13}$ ,  $1.226 \times 10^{-13}$  and  $1.310 \times 10^{-14} \text{m}^2/\text{sec}$ , respectively. When the as-pressed density of bentonite increased from 1.8 to 2.0 g/cm<sup>3</sup>, the apparent diffusivity of Cs-137 decreased by quarter. In the case of bentonite heat-treated to 150°C, no significant change in diffusivity was observed, which showed the possibility that the domestic bentonite could be used as a chemical barrier to retard the radionuclide migration at below 150°C. From the calculated pore and surface diffusivity, the surface diffusion due to the concentration gradient of radionuclide sorbed on the solid phase was found to dominate greatly in total transport process.

*Key words* : Radionuclide diffusion, Bentonite, Waste disposal, Diffusivity.

### INTRODUCTION

Bentonite has been favoured in many countries for the radioactive waste disposal, either as a backfill or buffer material in underground repository, because bentonite has a low permeability to prevent the intrusion of groundwater toward the waste

package and a high sorptionability to slow the movement of released radionuclides when the waste package is breached. In the case of high-level waste(HLW) or spent fuel generating decay heat, however, the bentonite material has been evaluated as a type of compacted bentonite or mixture of bentonite and sand or crushed rocks to

improve its mechanical properties such as low thermal conductivity and bearing capacity[1-4]. To assess the performance of bentonite buffer used as the buffer materials in the spent fuel repository, the physical and chemical parameters of bentonite relevant to the radionuclide migration within the buffer under the expected conditions should be determined. In general, it is believed that the radionuclide migration in well compacted bentonite is governed by diffusion mechanism because of its low hydraulic conductivity of  $10^{-12} \sim 10^{-14}$  m/sec [5-7]. Many researchers have studied on the diffusion behavior of radionuclides in compacted bentonite[8-15]. But the bentonite material investigated in those papers are of sodium type, whereas in Korea available bentonite is mostly of calcium type.

In this study, to provide the basic data for assessing the applicability of domestic bentonite collected from the bentonite mining area (Yang Buk 4) as a radiological barrier in the spent fuel repository, the diffusion coefficients of Co-60, Sr-85, and Cs-137 in compacted domestic bentonite were measured. In order to investigate the compaction density and the thermal effects of bentonite on the diffusion, the apparent diffusivities of Cs-137 in compacted domestic bentonite were measured. Finally, from the measured diffusivity of anion, Cl-36, the diffusion mechanism of cationic radionuclides interested were analysed.

## DIFFUSION THEORY

The diffusion of radionuclide in a porous

medium is described by a phenomenological analogy to Fick's law and, in one dimension, is given by

$$\partial C / \partial t = D_a (\partial^2 / \partial X^2) \dots\dots\dots(1)$$

where  $C$  is the radionuclide concentration,  $t$  is time,  $x$  is the distance that the radionuclide has diffused and  $D$  is the apparent diffusivity of radionuclide in the porous medium and independent of concentration. When radionuclide migrate through the compacted bentonite, a sorptive porous medium, the overall radionuclide concentration in the porous medium would be considered as the sum of two concentrations: one dissolved in liquid phase,  $C_L$  and the other sorbed on the surface of solid phase,  $C_s$ . Here  $C_L$  would be related to only macropore water in which the dissolved radionuclide, which was not sorbed yet, or non-sorbing radionuclide is contained. And, if the sorption reaction between both liquid and solid phases is reversible in equilibrium state,  $C_s$  could be explained by the distribution coefficient,  $K_d$ , defined as the relative ratio of the radionuclide concentration bound to the solid phase to the concentration in liquid phase. Thus,

$$C = C_L + C_s = \theta_p C_p + K_d \cdot \rho \cdot C_p \dots\dots\dots(2)$$

where  $\theta_p$  is the volume fraction of macropore water in saturated bentonite,  $C_p$  is the radionuclide concentration in pore water and  $\rho$  is the dry bulk density.

According to Eq.(2), two diffusion processes, pore diffusion with a constant of  $D_p$  and surface diffusion with a constant of

$D_s$ , could take place due to the respective concentration gradient. When Eq. (2) is substituted into Eq. (1), therefore, the apparent diffusivity,  $D_a$ , could be expressed as :

$$D_a = \frac{D_p + (K_d \cdot \rho) D_s / \theta_p}{1 + K_d \cdot \rho / \theta_p} \dots\dots\dots (3)$$

where,  $D_p$  is the pore diffusivity of radionuclide and  $D_s$  is the surface diffusivity of radionuclide in compacted bentonite.

Assuming that there is no surface diffusion as in Trostenfelt's and other researchers' studies[10-11], Eq. (3) becomes

$$D_a = D_p / (1 + K_d \cdot \rho / \theta_p) \dots\dots\dots (4)$$

In this expression, the pore diffusivity,  $D_p$ , of radionuclide in compacted bentonite is commonly related to the diffusivity of radionuclide in free water,  $D_w$ , as[15] :

$$D_p = D_w \cdot f \dots\dots\dots (5)$$

where  $f$  is the geometrical factor that accounts for a tortuous path around the solid particles or the pores in the porous medium. And the values of  $D_w$  for radionuclides are usually available in the literature [19].

If  $x$  is the diffusion distance of radionuclide from the source layer and  $2d$  is total length of the diffusion medium, the initial and boundary conditions to apply Eq. (1) to the experimental diffusion system applied in this study are  $\partial C / \partial X(o, t) = 0$  and  $C(\pm d, t) = 0$ , respectively. The mathematical solution[16] of Eq. (1) becomes

$$C(x, t) = \frac{M \cdot \exp[-x^2 / (4D_a t)]}{2(\pi D_a t)^{1/2}} \dots\dots\dots (6)$$

where,  $C$  is the concentration of diffusing radionuclide at distance  $x$  (counts/m<sup>3</sup>),  $M$  is the total amount of radionuclide in source layer per unit area normal to the direction of diffusion (counts/m<sup>2</sup>),  $D_a$  is the apparent diffusivity (m<sup>2</sup>/sec),  $x$  is the distance from a source layer in the direction of diffusion (m) and  $t$  is the diffusion time after introduction of source layer (sec).

## EXPERIMENTAL

### Materials

Bentonite sample used was collected from Yun-Il mining area (Yang-Buk 4). The air-dried sample was ground to pass 60 mesh sieve and some was heated in an oven at temperatures within 100~150°C for 24 hours. The solution used was also the same synthetic groundwater as in the previous study[17, 18].

In order to make a thin source layer, a nylon net, 2cm in diameter, of about 0.2 mm mesh which has negligible sorption effect, was covered with the dough of synthetic groundwater and bentonite. After that, it was slowly heated under an IR-lamp. Its final dimensions were about 0.2 mm thick and 2 cm in diameter. About 1 ml of stock solution containing radionuclide was dropped onto the thin bentonite-plate and slowly evaporated under an IR-lamp. Thus, the thin source layer of Co-60, Sr-85, Cs-137 and Am-241 were prepared, respectively.

### Apparatus and procedures

The experimental equipments used are illustrated in Fig. 1~3, of which the designs are similar to the ones reported by Torstenfelt and co-workers[10,11]. The air-dried bentonite was placed in a sample holder, which was fixed between a sample guide and a base plate as shown in Fig. 1(a). Using a conventional press, the bentonite sample was compacted to the desired as-pressed density by a plunger. The final dimensions of the cylindrical compacted bentonite prepared by means of pressing device were 2 cm long and 3 cm in diameter. As shown in Fig. 1(b), the two types of sample holder, A and B, were prepared to fit well and to make one set of diffusion cell unit as in Fig. 2. After pressing, both the porous metal filters whose pore sizes are  $2\mu\text{m}$  were placed directly on both ends of sample body being held in sample holder A and B. The filters were held in position by top and bottom plates in which a hole of 4 mm is prepared for saturation and the sample holder was closed by bolting as shown in Fig. 1(b). After that, the sample holder was immersed in synthetic ground water for saturation for 20 days.

After the saturation of compacted bentonite, the bottom plate of sample holder A and the top plate of sample holder B were removed. The thin source layer was put on the open surface of compacted bentonite contained in the sample holder B, and the sample holder A was put on the top of it, which is denoted as a diffusion cell unit in Fig. 2. This diffusion cell unit was immersed in the synthetic ground water.

After an appropriate diffusion time, the diffusion cell unit was opened and fit to the sample extruding device as shown in Fig. 3. This device consists of three parts: plunger, screw and plunger & screw holder. The plunger was attached to the screw and functioned to extrude the compacted bentonite by the working of screw. Especially, its bottom section was designed to prevent axial rotation of the compacted bentonite during extrusion by giving a tapered end. The compacted bentonite is gradually pressed out of the sample holder, 0.5 mm at a quarter revolution of screw and its corresponding portion was removed with a sharp blade.

Finally, the radioactivity of radionuclide diffused in each individual section was measured to obtain the concentration profiles of radionuclide diffused into the bentonite sample.

### Determination of diffusivities

The apparent diffusivity of radionuclide was determined by fitting the concentration profile of each radionuclide to a curve derived from Eq. (6).

The apparent diffusivity of  $\text{Cl-36}$ , whose sorption onto bentonite is negligible, was also determined to obtain the  $f$  value of the bentonite compacted to  $1.8\text{g}/\text{cm}^3$ . From the knowledge of  $f$  and  $D_w$  of radionuclide obtained from Nernst and Haskell equation [19],  $D_p$  of radionuclide was calculated by Eq. (5). Finally,  $D_s$  of radionuclide was obtained by subtracting  $D_p$  from  $D_a$ .

### Measurement of volumetric water content

Prior to introducing the source layer into

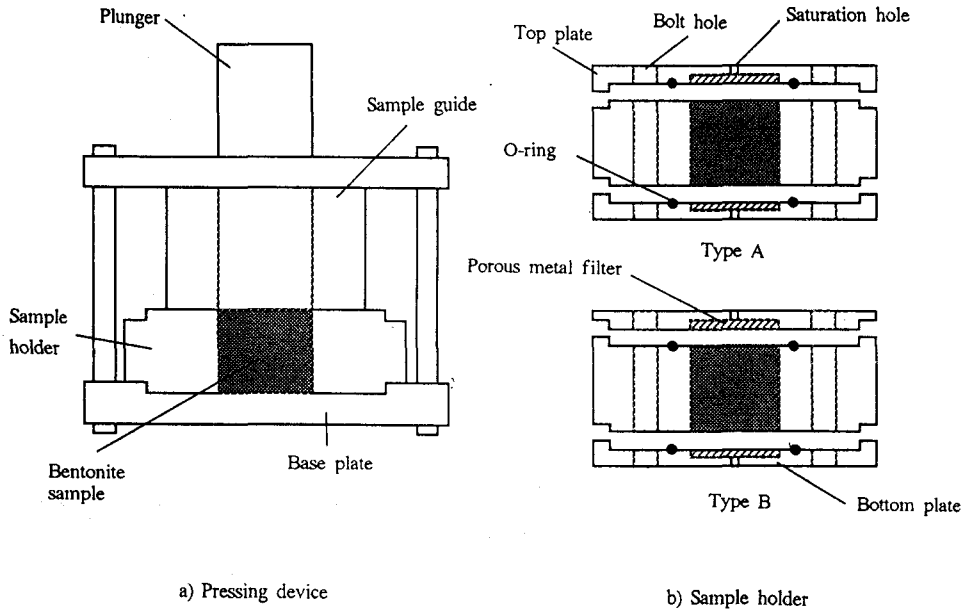


Fig. 1. Pressing device and sample holder.

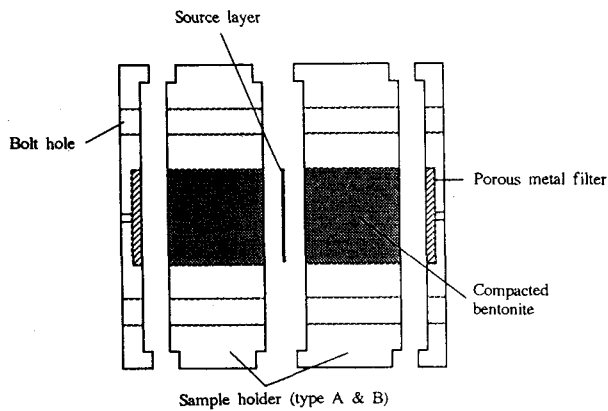


Fig. 2. Diffusion cell unit.

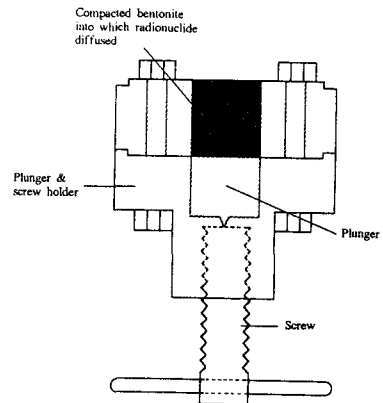


Fig. 3. Sample extruding device.

the middle of compacted bentonite sample, the water content of compacted bentonite sample was measured as a volumetric water content to identify its saturation for given saturation time. The saturated sample was sliced into thin sections of constant thickness. When the volume of section sliced was  $V_s \text{ cm}^3$ , the weight before drying was  $W_w$  and the weight after drying was  $W_d$ , the volumetric water content was calculated as  $[(W_w - W_d) / V_s] 100 (\%)$ .

## RESULTS AND DISCUSSION

### Water content and homogeneity of sample body

Fig. 4(a) shows the water content distribution of compacted bentonite held in sample holder with the longitudinal distance from the interface between filter and bentonite sample after saturation of 21 days. And Fig. 4(b) shows the water content distribution of compacted bentonite held in diffusion cell unit with the longitudi-

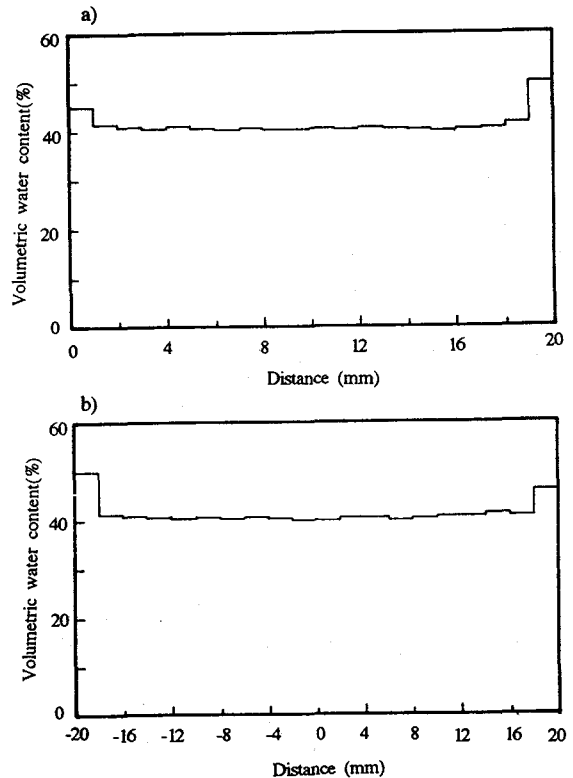


Fig. 4. Volumetric water content of compacted bentonite contained in a sample holder for 21 days (a) and diffusion cell unit for 71 days (b) (As-pressed density =  $1.8 \text{ g/cm}^3$ , Porosity = 43 %).

Table 1. Physical and chemical parameters for diffusion experiments.

Unit No.	As-pressed density ( $\text{g/cm}^3$ )		Porosity <sup>a)</sup> (%)	Water Content (wt.%)	Radionuclide	Kd ( $\text{ml/g}$ )
	Air dry	Oven dry				
1	1.77	1.59	40.98	$10.56 \pm 0.02$	Sr-85	$1345 \pm 23$
2	1.74	1.52	43.58	$12.60 \pm 0.06$	Cs-137	$5920 \pm 746$
3	1.99	1.78	33.9	$12.60 \pm 0.06$	"	"
4 <sup>b)</sup>	1.79	1.59	40.98	$11.10 \pm 0.03$	"	$7852 \pm 998$
5 <sup>c)</sup>	1.78	1.67	38.01	$6.20 \pm 0.01$	"	$10249 \pm 523$
6	1.74	1.52	43.58	$12.60 \pm 0.06$	Co-60	$7945 \pm 215$
7	1.75	1.53	43.54	$12.60 \pm 0.06$	Am-241	$54472 \pm 4056$
8	1.76	1.57	41.58	$10.53 \pm 0.02$	Cp-35	0

a) The particle density of Dong-Hae A is measured to be  $2.71 \pm 0.02 \text{ g/cm}^3$ .

b) Bentonite was heat-treated at  $100^\circ\text{C}$  before compaction.

c) " "  $150^\circ\text{C}$  " "

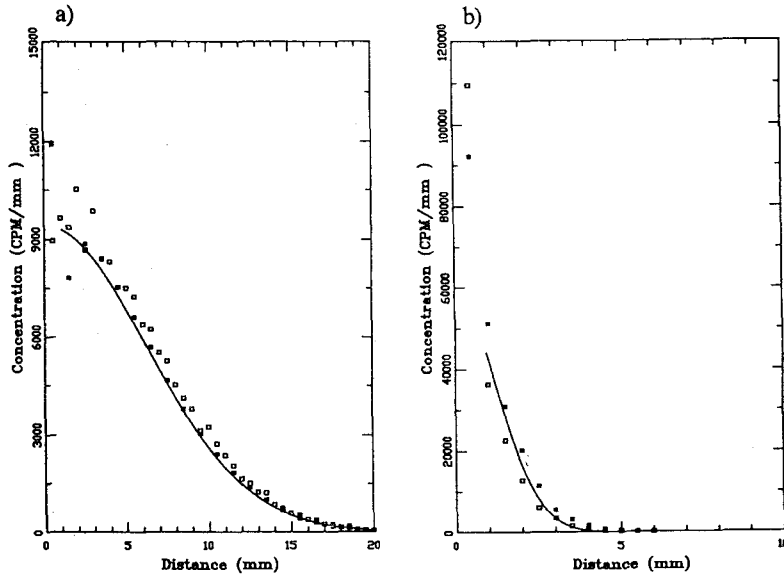


Fig. 5. Diffusion of Sr-85 (a) and Co-60 (b) in compacted bentonite (As-pressed density=18 g/cm<sup>3</sup>, Diffusion time=21 days(a) and 71 days(b)).

nal distance after diffusion time of 71 days under the assumption that the source layer is introduced into the middle of diffusion cell after saturation time of 21 days. Comparing the porosity of compacted bentonite, 42%, and the volumetric water contents of sections sliced, as a whole, the 94~97% of total pores in sample were found to be occupied by water. However, the higher volumetric water content than the porosity in both ends of compacted bentonite may be due to the excess water existing in porous filters of both sides when the diffusion cell unit was opened.

#### Diffusion of radionuclides

Physical and chemical parameters for the diffusion experiments are listed in Table 1.

As is seen in this table, the as-pressed densities of most samples are about 1.8 g/cm<sup>3</sup>, except for the sample of unit No.4 to investigate the effect of compaction density on the diffusion. And the diffusion cell unit No.5 and 6 are to investigate the effects of heat-treated bentonite on the diffusion. The distribution coefficients of radionuclides were measured by batch method in previous study[17, 18].

Fig. 5 to 8 show the radionuclide concentration profiles in the 20 mm long cylindrical bentonite samples. The solid lines in these figures show the concentration profiles calculated from Eq.(6), which gives the best fit to the experimental data with an respective apparent diffusivity,  $D_a$ . Fig. 8 shows that Am-241 rarely migrates through bentonite for 100 days. In Fig. 9, a straight

line plotted as  $\ln C$  versus  $x^2$  is illustrated as the measured concentration profile of  $\text{Cs-137}$  by applying natural log to Eq. (6).

In the vicinity of source layer, to about 2 mm, however, the calculated concentration profiles are significantly lower than the

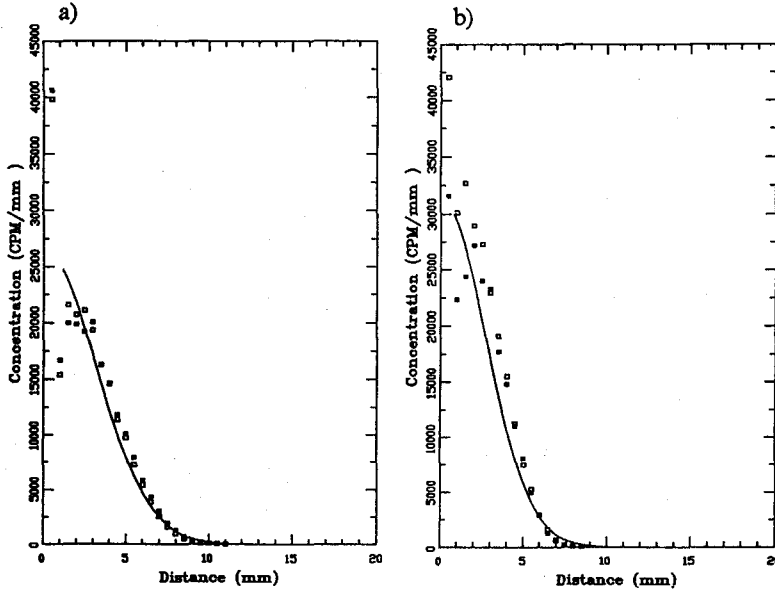


Fig. 6. Diffusion of  $\text{Cs-137}$  in compacted bentonite (As-pressed density = 1.8 (a) and 2.0  $\text{g/cm}^3$  (b), Diffusion time = 71 days).

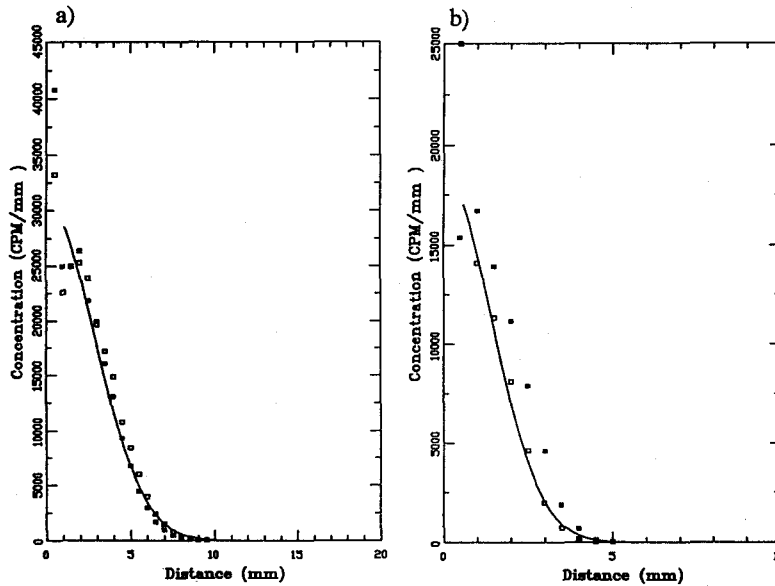


Fig. 7. Diffusion of  $\text{Cs-137}$  in compacted bentonite heat-treated at 100 (a) and 150 °C (b) (As-pressed density = 1.8  $\text{g/cm}^3$  and diffusion time = 71 days).



measured ones. This phenomena may be due to the experimental conditions being contrary to the theoretical assumptions :

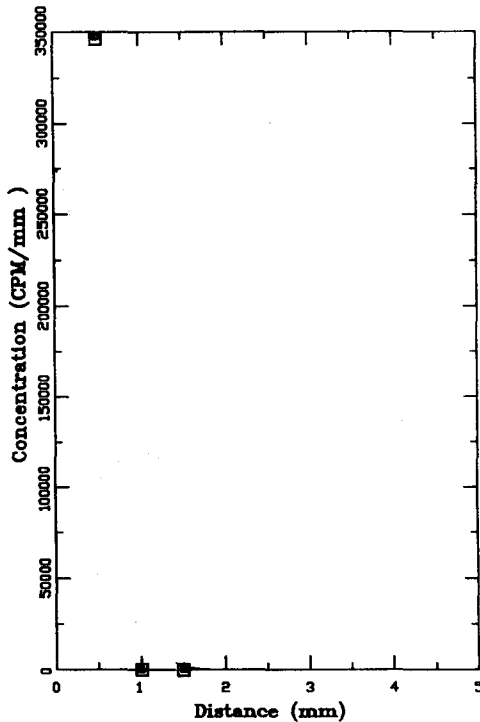


Fig. 8. Diffusion of Am-241 in compacted bentonite(As-pressed density=1.8 g/cm<sup>3</sup> and diffusion time=100 days).

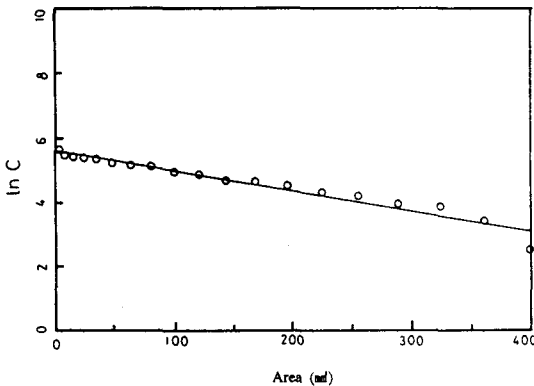


Fig. 9. Diffusion of Cl-36 in compacted bentonite(As-pressed=1.8 g/cm<sup>3</sup> and diffusion time=5 days).

independence of Kd-value on concentration. In the previous study[17], the maximum amount of Sr-85, Cs-137 and Co-60 sorbed onto Dong-Hae A sample was found to appear in the range of radionuclide concentration from 10<sup>-7</sup> to 10<sup>-5</sup> mol/l. Considering the fact that the radionuclide of 1.5~2.0μCi contained in the source layer was introduced into the bentonite with the water content of about 0.3ml/g, the radionuclide concentration introduced is evaluated to be about 10<sup>-3</sup> ~10<sup>-4</sup>mol/l. Therefore, some amount of radionuclide, which were not sorbed onto bentonite, could have existed in pore water.

In table 2, the apparent diffusivities determined from the measured concentration profiles of radionuclides are listed. The apparent diffusivities of Sr-85, Cs-137, Co-60, Am-241 and Cl-36 in compacted domestic bentonite with as-pressed density of about 1.8 g/cm<sup>3</sup> were measured to be 1.073×10<sup>-11</sup>, 6.705×10<sup>-13</sup>, 1.226×10<sup>-13</sup>, 1.310×10<sup>-14</sup> and 9.490×10<sup>-11</sup> m<sup>2</sup>/sec, respectively. These values are in good agreement with those reported by other workers[9,11,14,15] using a similar diffusion system. In this table, the measured apparent diffusivities of radionuclides could be arranged with the order of magnitude of distribution coefficient. These results may be understood from the evaluation of pore and surface diffusivities.

The values of D<sub>w</sub> were calculated to be the order of 10<sup>-9</sup> m<sup>2</sup>/sec and the value of f for the bentonite sample compacted to 1.8 g/cm<sup>2</sup> was estimated to be 0.048 by using Eq.(3) and (5). As described in Eq.(5), the pore diffusivities of radionuclides were

Table 2. Measured apparent diffusivities in domestic compacted bentonite.

Unit No. for experiment	Radionuclide	Important <sup>a)</sup> parameter	Kd · ρ ml/g	D <sub>a</sub> · 10 <sup>12</sup> m <sup>2</sup> /sec	Reference Data 10 <sup>12</sup> m <sup>2</sup> /sec
1	Sr-85	Density, 1.8 g/cm <sup>3</sup>	2139	10.73	10~48
2	Cs-137	Density, 1.8 g/cm <sup>3</sup>	8998	0.6705	0.9~2.5
3	"	Density, 2.0 g/cm <sup>3</sup>	10538	0.1669	0.5
4	"	Density, 1.8 g/cm <sup>3</sup>	12487	0.8467	-
5	"	Heat-treatment at 100°C Density, 1.8 g/cm <sup>3</sup>	17116	0.6045	-
6	Co-60	Heat-treatment at 150°C Density, 1.8 g/cm <sup>3</sup>	12076	0.1226	0.02~0.07
7	Am-241	Density, 1.8 g/cm <sup>3</sup>		0.0131	0.02~0.04
8	Cl-36	Density, 1.8 g/cm <sup>3</sup>		94.90	92

a) Important parameters according to experiment.

calculated by combining the value of  $f$ , 0.048, and the respective  $D_w$ .

The surface diffusivities of radionuclides were estimated by subtracting the calculated pore diffusivities from the measured apparent ones. The comparison of the measured apparent diffusivities and the surface diffusivities shows that the total transport rates of radionuclides in compacted bentonite depend on the surface diffusion. These results are similar to Eriksen's notes [15].

When the as-pressed density of compacted bentonite increased from 1.8g/cm<sup>3</sup> to 2.0g/cm<sup>3</sup>, the apparent diffusivity of Cs-137 decreased significantly to a quarter of the previous value. This is due to the tortuous paths and the sorption capacity increases and porosity decreases as the compaction density increases. In table 3, the apparent diffusivities of Cs-137 with varying the as-pressed density and the temperature of heat-treatment are listed. In the case of heat-treated bentonite with the as-pressed

density of 1.8g/cm<sup>3</sup>, no significant change in the apparent diffusivity over the range of room temperature to 150°C was observed in spite of increasing in Kd-values with temperature.

## CONCLUSIONS

The apparent diffusivities of Sr-85, Cs-137, Co-60, Am-241 and Cl-36 were measured to be  $1.073 \times 10^{-11}$ ,  $6.705 \times 10^{-13}$ ,  $1.226 \times 10^{-13}$ ,  $1.310 \times 10^{-14}$  and  $9.490 \times 10^{-11}$  m<sup>2</sup>/sec, respectively and could be arranged with the order of magnitude of their distribution coefficients, i.e. Cl > Sr > Cs > Co > Am. As the as-pressed density of bentonite sample increased from 1.8 to 2.0g/cm<sup>3</sup>, the apparent diffusivity of Cs-137 decreased by quarter. In the case of bentonite heat-treated to 150°C, no significant change in diffusivity was observed.

From the calculated pore and surface diffusivity of Sr-85, Cs-137 and Co-60, it was found that the surface diffusion due to

**Table 3. Calculated pore and surface diffusivities in domestic compacted bentonite.**(As-processed density = 1.8 g/cm<sup>3</sup>).

Radionuclide	D <sub>s</sub> 10 <sup>12</sup> m <sup>2</sup> /sec	Kd ρ θ <sub>p</sub>	f	D <sub>w</sub> <sup>a)</sup> 10 <sup>9</sup> m <sup>2</sup> /sec	D <sub>ac</sub> <sup>b)</sup> 10 <sup>12</sup> m <sup>2</sup> /sec	D <sub>p</sub> 10 <sup>11</sup> m <sup>2</sup> /sec	D <sub>s</sub> 10 <sup>12</sup> m <sup>2</sup> /sec	Reference data[18, 19]		
								m <sup>2</sup> /sec	m <sup>2</sup> /sec	m <sup>2</sup> /sec
	10.730	5499	0.048	0.78	0.0068	3.744	10.720	15	3.5	13.5~15
	0.671	21734	0.048	2.11	0.0047	10.128	0.666	2.5	9.5	0.5~2.1
	0.123	29170	0.048	1.25	0.0021	6.000	0.120	0.02	-	0.039

a) The diffusivity of radionuclide in free water, which is calculated by Nernst and Haskell equation [25] as :

$D_w = \frac{RT}{F^2} \cdot \frac{1/n_+ + 1/n_-}{1/\lambda_+ + 1/\lambda_-}$ , where R is the gas constant, J/g-mol K=8.314, T is the temperature, K,  $\lambda_+/\lambda_-$  is the limiting (zero concentration) ionic conductance, A/[cm<sup>2</sup> · (V/cm) (g-equiv/cm<sup>3</sup>)],  $n_+/n_-$  is the valence of ion, and F is the faraday constance, c/g-equiv.

b) The calculated apparent diffusivity of radionuclide by using Eq. (4).

the concentration gradients of radionuclide sorbed on the bentonite particle is a dominating transport process of radionuclide in compacted bentonite. One of the ways to prevent the dorminating surface diffusibility of radionuclide sorbed is thought to increase the compaction density of bentonite buffer.

Some data obtaned as the results would contribute to the engineering parameters to design the waste package and engineered barrier satisfying the safety requirements. Especially, the effects of heat-treated bentonite on the diffusion would be based on the establishment of thermal limit from the view point of thermal stability of bentonite buffer.

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## 압축 국산 벤토나이트 내에서 방사성 핵종의 확산이동

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### 요 약

압축된 국산벤토나이트에서 Sr-85, Cs-237, Co-60 및 Am-241의 확산연구를 수행하였다. 본 실험에서는 원통형으로 압축된 벤토나이트 시료의 중앙부에서 축방향으로 방사성 핵종의 확산이동이 이루어지도록 하여 각 방사성핵종의 확산계수를 측정하였다. 그리고 벤토나이트의 열처리 온도와 압축밀도가 확산에 미치는 영향 등을 분석하였다.

Sr-85, Cs-137, Co-60 및 Am-241의 겔보기 확산계수는 각각  $1.07 \times 10^{-11}$ ,  $6.705 \times 10^{-13}$ ,  $1.226 \times 10^{-13}$ ,  $1.310 \times 10^{-14}$  m<sup>2</sup>/sec로 측정되었다. 그리고 시료의 압축 밀도를 1.8 g/cm<sup>2</sup>에서 2.0 g/cm<sup>2</sup>으로 증가시켰을 때, Cs-137의 확산계수는 약 1/4로 감소되어 나타났다. 반면, 열처리된 벤토나이트의 경우에는 확산계수가 크게 변하지 않았는데, 이는 150°C 이하의 온도에서는 국산 벤토나이트가 방사성핵종의 이동을 지연시킬 수 있는 화학적 방벽으로서 사용할 수 있다는 가능성을 보여준 것이라 생각된다. 그리고 음이온 Cl-36의 확산계수를 이용하여 도출한 각 방사성핵종의 공극확산계수와 표면확산계수를 측정한 겔보기확산계수와 비교해 볼 때, 전체 방사성 핵종의 확산이동에 있어서 표면확산이동이 지배적인 것으로 나타났다.