

## Experimental Study on the Hydrodynamic Dispersion of Contaminants in Geologic Media : Adsorption and Diffusion of Sr and Cr-EDTA in Granitic Rocks

수리지질계에서 지질매체에 따른 오염물질의 수리분산에 관한 실험적 연구  
: 화강암질암에서 Sr과 Cr-EDTA의 흡착 및 확산에 관한 연구

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**Abstract** : To investigate the migration behavior of contaminants in rocks, adsorption and diffusion experiments for Sr as a sorbing contaminant and for Cr-EDTA as a non-sorbing contaminant were carried out on granitic rocks. The Sr adsorption on separated minerals and crushed rocks tends to slightly increase with increasing pH. It also greatly decreases with the increase of ionic strength in NaCl solution. Among separated minerals, biotite and sericite have adsorbed much more amount of Sr than other rock-forming minerals, such as quartz, plagioclase, and potassic feldspar, because the specific surfaces and cation exchange capacities of phyllosilicates are generally much greater than those of the other rock-forming minerals. The intrinsic diffusion coefficients of Cr-EDTA for granitic rocks differ little from those of Sr. This indicates that they are independent of water-rock interactions. Experimental data show that the intrinsic diffusion coefficients are positively correlated with the porosities of the rocks. They are close to the theoretically predicted values, especially in pre-steady state diffusion region, with the increase of rock sample thickness.

**요 약** : 수리지질계에서 지질매체에 따른 오염물질들의 분산 거동을 알아보기 위하여, 흡착 오염물질로서 Sr과 비흡착 오염물질로서 Cr-EDTA를 대상으로 화강암질암에 대한 흡착 및 확산실험을 수행하였다. 분리된 광물 및 파쇄된 암석에 대한 Sr 흡착은 pH가 증가함에 따라 더불어 증가됨을 보여준다. 또한, 용액내의 NaCl의 농도가 증가하면서 이온세기가 높아지게 되면, 시료에 흡착되는 Sr의 양은 급격히 감소하였다. 분리된 광물 시료 중에서는 흑운모와 견운모가 석영, 사장석 및 칼리장석과 같은 다른 조암광물에 비하여 Sr을 훨씬 많이 흡착하였는데, 이는 일반적으로 층상 규산염광물들이 다른 조암광물들보다 비표면적과 양이온 교환능력이 훨씬 크기 때문인 것으로 판단된다. 동일한 암석인 경우, 비흡착 물질과 흡착 물질간의 고유확산계수 값에는 거의 차이가 없었다. 이는 암석의 고유확산계수가 암석과 유체 상호간의 화학반응에 거의 영향을 받지 않음을 암시한다. 또한 암석의 고유확산계수는 공극률이 커질수록 큰 값을 보여준다. 암석의 두께가 두터워질수록 전정상정류구간(pre-steady state region)에서의 실험자료와 이론적으로 계산된 곡선 사이의 불일치 정도는 감소하는 경향을 보여주는데, 이는 사공극 확산효과(dead-end pore diffusion effect)가 암석의 두께가 두꺼워질수록 줄어들음을 의미한다.

### INTRODUCTION

The critical concentration values of contaminants in biosphere depend on not only groundwater flow but also the interaction between contaminants and rocks.

The transport of contaminants is affected by various parameters, such as distribution coefficient, diffusion coefficient, and

dispersivity. The parameters can be obtained from adsorption and diffusion experiments in field and laboratory.

In the study, in order to investigate the behavior of contaminants in granitic rocks, adsorption and diffusion experiments for Sr as a sorbing contaminant and Cr-EDTA as a non-sorbing contaminant were carried out on granitic rocks as a function of contaminant concentration, pH, and ionic strength.

### EXPERIMENTAL PROCEDURE

#### Samples used in experiments

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**Mugeug Granodiorite:** The granodiorite is distributed near Mugeug Gold Mine located at Yonggyeri, Geumwang-eup, Eumsung-gun, Chungcheongbuk-do (Cheong *et al.*, 1976). The rock shows medium-grained equigranular but locally porphyritic texture with the phenocrysts of pinkish potassic feldspar. The texture changes slightly in place without a wide variation in whole rock compositions. The rock mainly consists of quartz, potassic feldspar, plagioclase, biotite, hornblende, and the accessory minerals, such as apatite, sphene, zircon, and opaque minerals. Biotite was slightly altered to chlorite along its cleavage and often replaced by sericite. Hornblende shows poor cleavage and occurs as subhedral to anhedral crystals and encloses fine-grained pyroxenes as a remnant. Sphene occurs as euhedral crystals and it was commonly enclosed by hornblende and/or biotite. Mafic minerals such as biotite and hornblende and opaque minerals constitute 16 vol.% of the rock.

**Naeduckri Granite:** The Naeduckri granite is distributed in the whole district of Naeduckri, Sangdong-eup, Youngwol-gun, Kangweon-do. The rock is tourmaline-bearing granite, which is coarse-grained and equigranular. The rock mainly consists of quartz, potassic feldspar, plagioclase, tourmaline, muscovite, and/or biotite and the accessory minerals, such as apatite, sphene, zircon, and opaque minerals. Both of muscovite and biotite were partly altered to sericite.

### Adsorption Experiments

The rock samples of Mugeug granite and Naeduckri granite were crushed in an agate mortar to avoid iron contamination, and sieved. The crushed rock samples with the size of 0.125 to 0.177 mm and 0.25 to 0.50 mm in diameter were used in adsorption experiments. Minerals were separated from one another by magnetic isodynamic separation and hand extraction methods. The groundwater used in the study was taken from Mt. Kwanak located at Sillimdong, Kwanakgu, Seoul. The chemical composition of the groundwater is given in Table 1. The crushed and sieved rock samples were contacted with the groundwater by washing of three or four times and then dried at 105°C for 12 hours in an oven. The Sr solution samples, where the concentration of Sr ranges from 5.71 to 571 mg/l were prepared using Sr(NO<sub>3</sub>)<sub>2</sub>. One gram of crushed rock samples was equilibrated with 4 ml of the each Sr solution at 24±1°C for five days in a glass vial. Subsequently,

**Table 1.** Chemical composition of the groundwater used in adsorption experiments.

Solute	Na	Mg	K	Ca	Cl
Conc.(ppm)	6.6	1.1	1.3	0.5	4.5

※ The concentrations of cations were determined by AAS and that of chloride by ion selective electrode. pH was 6.22.

aqueous phase was separated from solid phase by a centrifuge, and the concentrations of Sr in the aqueous phase were measured with atomic absorption spectrometry (AAS).

The pH and ionic strength of solutions were adjusted using diluted NaOH (or HCl) and NaCl solutions, respectively. To attain the equilibrium between solid and liquid phases, crushed rock samples and separated minerals were mixed with the solution added Sr to deionized water. The solid to liquid ratio was 1/10 (g/ml) and the initial Sr concentration of the solution was 10 mg/l. After the samples mixed with the Sr solution were centrifuged, the upper part of liquid phase was taken and analyzed using AAS for Sr concentration. Blank samples were run as well to measure the adsorption on the walls of vials. The experimental conditions are summarized in Table 2.

### Diffusion Experiments

The method used for diffusion experiments is in principle the same as that described previously by Bradbury and Green (1985, 1986), Lever *et al.*, (1985), and Skagius and Neretnieks (1982, 1986). The diffusion cell apparatus consists of a holder made of the acrylic plate of 8 mm in thickness. The holder is divided into two half-cells by a rock slab fixed by silicon glue in the center of the holder. One half-cell of the holder is a high concentration reservoir for solute and the other a measurement cell. Skagius and Neretnieks (1982) found that the diffusion of Cr-EDTA through silicon glue was so small as to be negligible.

The tracers used in diffusion experiments were Cr-EDTA and Sr. Cr-EDTA as a non-sorbing species, was chosen because its very weak sorption property, which minimizes any uncertainty due to the complex sorption process (Abelin and Neretnieks, 1981). Other non-sorbing species, which have been used in diffusion experiments by other investigators (Strickert *et al.*, 1980; Skagius and Neretnieks, 1984, 1986; Bradbury and Green, 1985; Lever *et al.*, 1985), are chloride, iodide, and uranin. Sr as a sorbing species was chosen because it has moderately sorbing feature (Keren and O'connor, 1983).

The reservoir was filled with the mixed solution of 0.1 M Cr-EDTA and 0.1 M Sr(NO<sub>3</sub>)<sub>2</sub>, whereas the measurement cell with deionized water. The solution sample of 5 ml was extracted every two days from the measurement cell, and then

**Table 2.** Conditions for adsorption experiments.

Solid phase	minerals and crushed rocks
Particle size	0.25~0.05 mm in diameter
Aqueous phase	deionized water
Solid/liquid ratio	1 g/10 ml
Initial strontium concentration	10 ppm
Temperature	24±1°C
Equilibration time	5 days
Analytical instrument	AAS and ion selective electrodes

the distilled water of 5 ml was added to the measurement cell to keep the constant volume. The concentrations of diffused Sr and Cr-EDTA in the measurement cell were determined with AAS.

## THEORY AND MECHANISM

### Adsorption

The ion exchange and adsorption of contaminants on minerals and rocks decrease the concentration of contaminants in groundwater. Among various chemical reactions affecting the migration rates of contaminants in groundwater, ion exchange appears to be the predominant process with respect to Sr. The degree of ion exchange is commonly represented by a distribution coefficient ( $K_d$ ), which describes the equilibrium partitioning of a contaminant between solid phase and aqueous phase. The migration rate of contaminants depends on the distribution coefficient, such that the greater distribution coefficient the slower migration rate.

The distribution coefficients used in models predicting the migration rates of contaminants require two major assumptions (Reynolds *et al.*, 1982): the reaction of solute with solid phase during transport maintains reversible equilibrium, and the ratio of the concentration of solute component in solid phase to that in liquid phase must remain constant.

Well known equilibrium adsorption isotherms are Freundlich isotherm and Langmuir isotherm, of which the latter was originally derived from the adsorption of gases by solids. Though the distribution coefficient of an element can be obtained from the linear portion of Langmuir isotherm, in many instances, experimental data do not well conform to Langmuir isotherm (O'Conner and Connolly, 1980; Skagius and Neretnieks, 1988). Accordingly, they can be fitted by the empirical relation known as Freundlich isotherm,

$$q = K C_e^N \quad (1)$$

where  $q$  is the mass of chemical species sorbed by solid phase, and  $K$  and  $N$  are Freundlich distribution coefficient and exponent coefficient, respectively.  $C_e$  is the dissolved species concentration in aqueous phase at equilibrium. The linear portion of Langmuir isotherm corresponds to Freundlich isotherm for  $N=1$ .

### Diffusion

When contaminants in groundwater are sorbed on or chemically react with fissure surfaces or micropore surfaces in the rock body, the diffusion process acts as a retarding mechanism for the contaminants.

The theory and experimental methods on the diffusion of dissolved chemical species have been described by previous in-

vestigators (Bradbury and Green, 1985; Skagius and Neretnieks, 1986). The diffusion equation can be expressed from Fick's second law:

$$\alpha \frac{\partial C}{\partial t} = D_i \frac{\partial^2 C}{\partial x^2} \quad ; \quad \alpha = \epsilon_{tot} + K_d \rho \quad (2)$$

where,  $D_i$  : intrinsic (effective) diffusion coefficient for  $i$  species

$\alpha$  : rock capacity factor

$\epsilon_{tot}$  : total porosity

$K_d$  : adsorption coefficient

$\rho$  : density of material

$D_i$  is related to the free water diffusion coefficient ( $D_0$ ) and the formation factor ( $\phi$ ).

$$D_i = D_0 \phi \quad (3)$$

The formation factor ( $\phi$ ) depends on various pore geometries and is often written as

$$\phi = \frac{\epsilon^* \delta}{\tau^2} \quad (4)$$

where  $\epsilon^*$  : through-transport porosity

$\delta$  : constrictivity

$\tau$  : tortuosity

The equation (2) can be solved for porous rocks with initial inlet concentration,  $C_0$  at  $x=0$  and outlet concentration,  $C_r$  ( $C_r \ll C_0$ ) at  $x=l$  in thickness of sample. From the equation (2), the total amount  $Q$  of a diffused species through porous rock after time  $t$  was solved by Skagius and Neretnieks (1986):

$$C_r = \frac{Q}{A l C_0} = \frac{C_i V}{A l C_0} \\ = \frac{D_i t}{l^2} - \frac{\alpha}{6} - \frac{2\alpha}{\pi^2} \sum_{n=1}^{\infty} \frac{(-1)^n}{n^2} \exp\left(-\frac{D_i n^2 \pi^2 t}{l^2 \alpha}\right) \quad (5)$$

where  $A$  and  $V$  are cross-section area and solution volume, respectively.  $C_r$  is the relative concentration of a diffused solute through the rock sample after time  $t$ .

For large times, the exponential terms in equation (5) can be negligible, and consequently, the asymptotic solution of equation (5) for the steady-state diffusion region is

$$C_r = \frac{D_i t}{l^2} - \frac{\alpha}{6} \quad (6)$$

From a plot  $C_r$  against  $t/l^2$ , both of  $D_i$  and  $\alpha$  can be calculated from the steady state diffusion region of the linear portion of the curve.

The intrinsic diffusion coefficient ( $D_i$ ) is independent of the sorption behavior of diffusing species. In contrast, the rock capacity factor ( $\alpha$ ) strongly depends on the chemistry of diffusing species and essentially determines the time scale to reach the steady state diffusion region in laboratory experiments.

In laboratory diffusion experiments, the following three particulars are not considered (Lever *et al.*, 1985): (1) the effect of lithostatic pressure, which may reduce porosity and diffusion coefficient, is not simulated, (2) the experiments are carried out on relatively small rock samples, in which the pore-connectivity may be greater than that in large intact rock mass, and (3) natural rock surfaces generally covered with weathering products such as clay-like minerals are not taken into account in the experiments.

**RESULTS AND DISCUSSION**

**Adsorption**

The results of the Sr batch experiments for Mugeug granodiorite and Naeduckri granite are shown in Figure 1. As stated above, the linear isotherm of Sr on a log-log plot can be interpreted by Freundlich isotherm. The distribution coefficients ( $K_d$ ) of Sr and N values for Mugeug granodiorite and Naeduckri granite are obtained from Figure 1 as 2.30 ml/g and 1.14 and 1.55 ml/g and 1.18, respectively. The difference in the distribution coefficients of Mugeug granodiorite and Naeduckri granite is probably due to the difference in mineral compositions. Modal analyses inform that biotite amounts in Mugeug granodiorite and Naeduckri granite are 14.5 and 1.4 vol. %, respectively. Biotite has large cation exchange capacity and specific surface area. According to Allard *et al.*, (1980), its cation exchange capacity and specific surface

area are about 17 meq/kg and 14 m<sup>2</sup>/g, respectively, of which the values are much higher than those of quartz and feldspar. The sorption capacities of these minerals are also proved by adsorption experiments for separated minerals and crushed rock samples (Figure 2).

The Sr adsorption on Mugeug granodiorite is about two times greater than that on Naeduckri granite (Figure 2). As mentioned above, the amount of Sr adsorbed on rocks depends mainly upon the mineral composition of rocks. The Sr

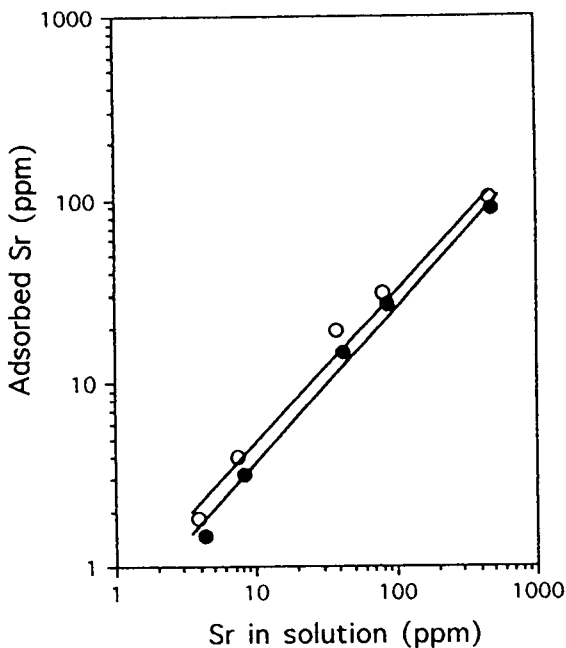


Figure 1. Sr adsorption isotherms for Mugeug granodiorite (closed circles ;  $K_d=2.30$  ml/g) and Naeduckri granite (open circles ;  $K_d=1.55$  ml/g).

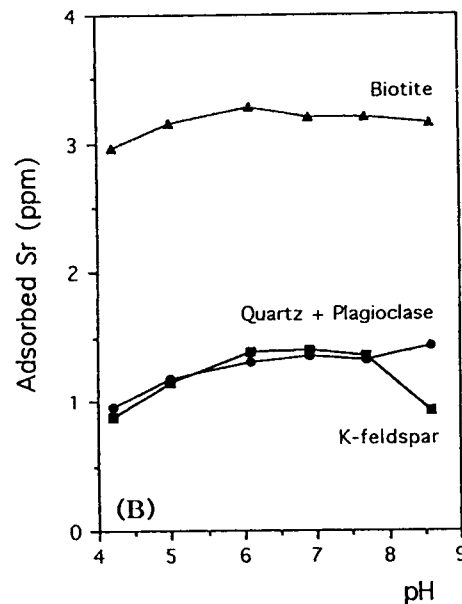
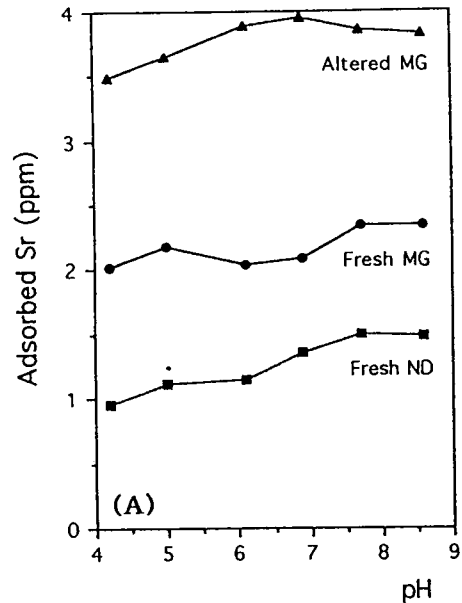


Figure 2. Sr adsorption on crushed rocks (A) and separated minerals (B) as a function of pH. All minerals were separated from Mugeug granodiorite. MG: Mugeug granodiorite, ND: Naeduckri granite.

adsorption on the hydrothermally altered Mugeug granodiorite is greater about two times than that on the fresh equivalent. This seems to be due to the presence of secondary alteration minerals such as sericite having high sorption capacity.

The Sr adsorption on the minerals in Mugeug granodiorite are shown in Figure 2. The sorption capacity of biotite is much higher than that of the other minerals. These ex-

perimental results coincide with those of Allard *et al.* (1982).

In general, adsorption increases as pH increases, but in Figure 3 the Sr adsorption on minerals and crushed rocks does not show distinct change with pH. These results show that the higher ionic strength causes the less adsorbed Sr, and that the Sr adsorption on samples is markedly depressed by the presence of sodium ion. Keren and O'Connor (1983) also indicated that the adsorption capacity of rocks decreased exponentially with the increase of ionic strength. This effect is known as "competition effect".

### Diffusion

All pores in rocks can be divided into two groups: through-transport pores and dead-end pores (Lever *et al.*, 1985; Skagius and Neretnieks, 1986). The dead-end pores are not necessarily to be closed at one end, as even the dead-end pores connected to through-transport pores at both ends have a no-flux boundary at some point along their length (Lever *et al.*, 1985). Although dead-end pores are of no importance for repository-scale migration, they have implications for laboratory-scale migration. The concentration-time curve for the diffusion of an element is divided into the pre-steady state diffusion region and steady state diffusion region. The steady state diffusion takes place only in the through-transport pores. The dead-end pores acting as sinks and/or sources of contaminants contribute mainly to the pre-steady state diffusion.

Intrinsic diffusion coefficient ( $D_i$ ) and rock capacity factor ( $\alpha$ ) are determined by a least square fit from the steady state diffusion region of the concentration-time curve and they are listed in Table 3. The intrinsic diffusion coefficients of Cr-EDTA and Sr in Mugeug granodiorite are  $7.63 \times 10^{-15}$  to  $4.75 \times 10^{-13}$  m<sup>2</sup>/s and  $1.61 \times 10^{-13}$  to  $4.59 \times 10^{-13}$  m<sup>2</sup>/s, respectively. The intrinsic diffusion coefficients for Cr-EDTA and Sr are similar. This indicates that there is no significant difference in intrinsic diffusion coefficients between non-sorbing and sorbing species in small-scaled laboratory experiments.

The increase in the thickness of rock slabs induces the increase of the formation factor ( $\phi$ ) expressing the pore geometry of rock (Table 3).

The experimental data for Cr-EDTA diffusion in Mugeug granodiorite clearly show pre-steady state (A) and steady state (B) diffusion regions on the theoretically calculated curve from equation (6) (Figure 4). In the thin rock sample of 0.24 cm in thickness (Figure 4), a discrepancy between the experimental data and the theoretically calculated curve exists in the pre-steady state diffusion region, i.e. the concentrations theoretically predicted are consistently lower than those experimentally measured. Whereas, this discrepancy decreases for the samples of 0.50 cm in thickness (Figure 5). Bradbury

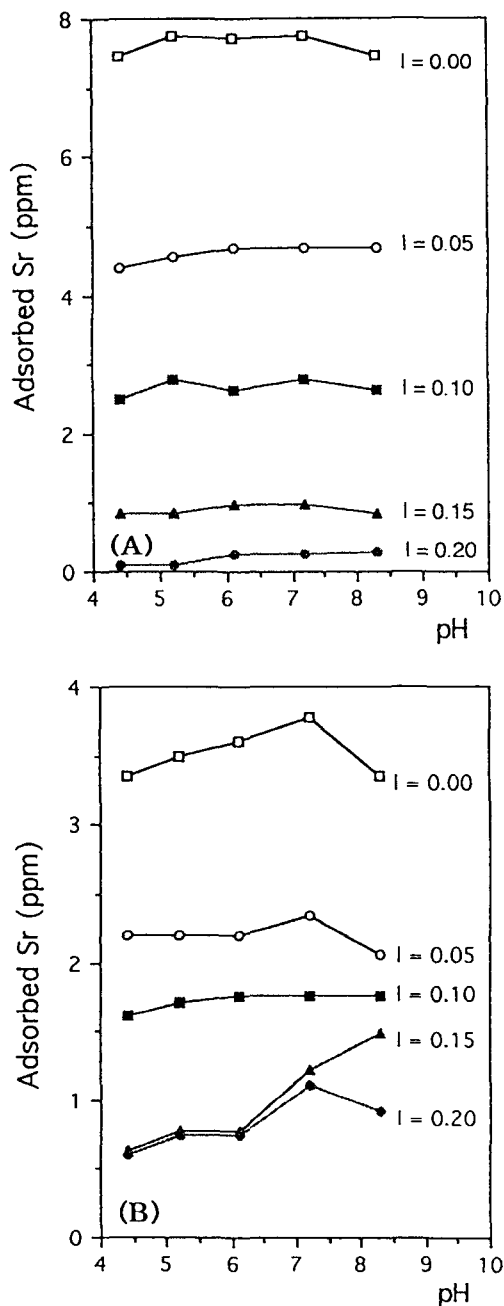
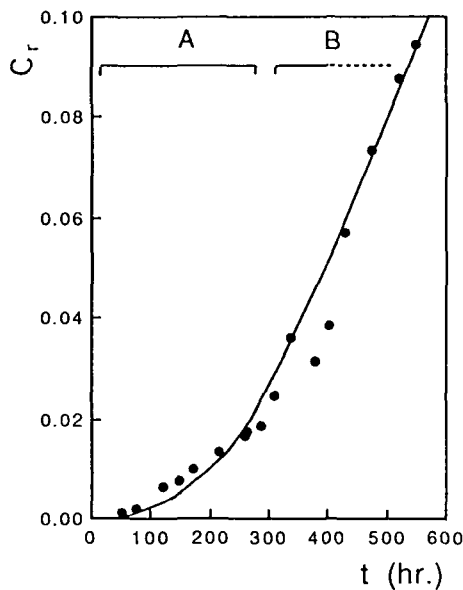


Figure 3. Sr adsorption on altered Mugeug granodiorite (A) and Naeduckri granite (B) as a function of pH and ionic strength(I) in NaCl solution.

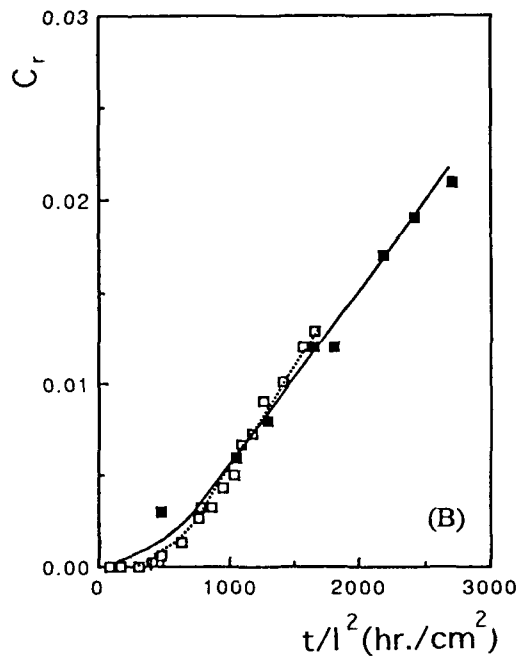
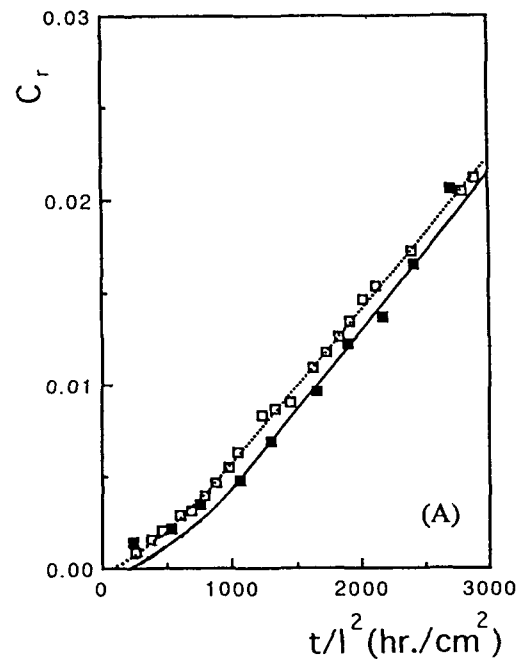
**Table 3.** Intrinsic diffusion coefficients ( $D_i$ ) and rock capacity factors ( $\alpha$ ) determined by experimental data.

Sample	Species	$D_i$ ( $10^{13} \text{ m}^2/\text{s}$ )	$\alpha$ (%)	Thickness (cm)	Formation Factor( $10^4$ )	Porosity (%)
Mugeug Granodiorite	Cr- EDTA	4.75	42.2	0.24	2.97	0.45-0.71
		2.40	2.49	0.30	1.50	
		2.29	1.49	0.50	1.43	
		0.76	2.11	0.52	0.48	
	Sr	1.61	2.69	0.30	1.24	
		3.33	4.06	0.55	2.56	
Naeduckri Granite	Cr- EDTA	18.0	1.62	0.50	11.3	0.98-1.74



**Figure 4.**  $C_r$  vs.  $t$  plot. Cr-EDTA diffusion in Mugeug granodiorite shows pre-steady state diffusion region (A) and steady state diffusion region (B). Solid line represents theoretically calculated curve. Closed circle represents the experimental data for the rock sample of 0.24 cm in thickness.

and Green (1985) and Lever *et al.* (1985) explained this phenomenon with dead-end pore model. According to them, the influence of dead-end pores on the pre-steady state diffusion region could be observed only in relatively thin samples (< 5 cm) and it was progressively less effective as the sample thickness increases. Thus, dead-end pores have little effect on the migration model of contaminants in the repository scale. Therefore, the simple diffusion equations derived from Fick's second law are adequate because the rock body used for waste disposal is much more greater than the rock slab samples used in experiments.



**Figure 5.**  $C_r$  vs.  $t/l^2$  plot showing Cr-EDTA diffusion curves (A) and Sr diffusion curves (B) for Mugeug granodiorite samples of different thicknesses. Closed and open squares represent the experimental data for two rock samples of 0.3 cm and 0.5 cm in thickness, respectively.

The diffusion curves of Cr-EDTA for each of two Mugeug granodiorite samples of 0.3 cm and 0.5 cm in thickness are shown in Figure 5 (A) and those of Sr in Figure 5 (B). Comparing Figure 5 (A) with Figure 5 (B), there is little or no difference in the slopes of the curves i.e. in the intrinsic diffusion

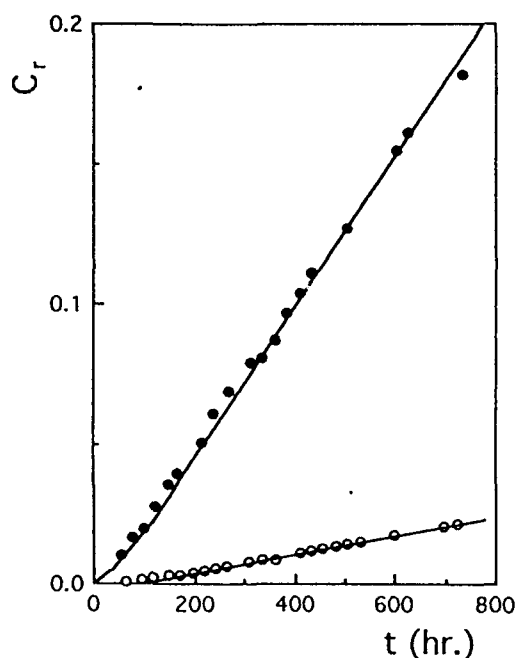


Figure 6.  $C_t$  vs.  $t$  plot showing Cr-EDTA diffusion in Mugeug granodiorite (open circles) and Naeduckri granite (closed circles) in same thickness.

coefficients of Cr-EDTA as a non-sorbing species and Sr as a sorbing species. This indicates that the intrinsic diffusion coefficients are independent of the sorption behavior of diffusing species.

Compared the diffusion curves of Cr-EDTA in Mugeug granodiorite with those in Naeduckri granite with the same thickness of 0.5 cm (Figure 6), the intrinsic diffusion coefficients of Naeduckri granite are about six times greater than those of Mugeug granodiorite. This may be due to the difference in porosities between the two rocks. The porosity of Naeduckri granite (0.98 - 1.74 %) is much greater than that of Mugeug granodiorite (0.45 - 0.71%). The results shown in Figure 6 indicate that the diffusion of species is significantly controlled by the porosity of rocks.

### CONCLUSIONS

Based on the experimental study on the adsorption and diffusion of Sr as a sorbing contaminant and Cr-EDTA as a non-sorbing contaminant in granitic rocks, we can conclude as follows:

1. The Sr adsorption on separated minerals and crushed rocks tends to slightly increase with increasing pH, but it does not show distinct change with pH in NaCl solution. This indicates that it greatly decreases with the increase of ionic strength in solution. The Sr adsorption on both of biotite and sericite is much greater than that on oth-

errock-forming minerals such as quartz, plagioclase, and potassic feldspar.

2. The significant discrepancy between the experimental data and the theoretically calculated curves in pre-steady state diffusion region decreases with the increase of rock thickness. This indicates that the effect of dead-end pores on diffusion process progressively decreases as the rock thickness increases.
3. There is little difference in the intrinsic diffusion coefficients of both of Cr-EDTA and Sr in granitic rocks. The intrinsic diffusion coefficients are independent of chemical reactions such as sorption. They are positively correlated with the porosities of rocks.

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