

A Study on the Underground Movement of Radionuclides(I)

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방사성핵종의 지하이동 연구

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Abstract

With regard to the radioactive waste disposal, adsorption properties and migration rates have been evaluated for Cs-137 and Sr-90 with the domestic clay sampled from Cnyang, Sanchong and Mooan. Sorption coefficients(Ksorp) were determined by batch experiments. The measured values of Ksorp were ranged from 8000 to 17,000 ml/gr for Cs-137 of 0.1 μ Ci/ml, and from 10,000 to 15,000ml/gr for Sr-90 of 0.1 μ Ci/ml. Remarkably, Mooan clay showed lower values of Ksorp than those of the others. This could be explained by the poor sorption capacity of the quartz found only in the Mooan clay.

For the quantitative analysis, sorption isotherm equations of Freundlich type were made with the obtained values of Ksorp

$$C_R = 18.0 C_A^{0.74} \quad \text{for Cs-137}$$

$$C_R = 0.84 C_A^{0.45} \quad \text{for Sr-90}$$

By introducing the BOX model combined with the above relationships, simulation of underground nuclide movement was carried out. The results showed that the domestic clays could be the effective backfill material for repositories.

요 약

방사성폐기물 처분과 관련하여 산청, 은양 그리고 무안에서 채취한 국산점토에 대한 Cs-137 및 Sr-90의 흡착특성과 이들 핵종의 점토층이동에 대하여 고찰하였다. 흡착분배계수(Ksorp)를 회분식 흡착 실험으로 결정한 결과 Cs-137의 경우 8,000-17,000ml/g 그리고 Sr-90의 경우 10,000-15,000ml/gr 범위의 값이었다. 이때 액상의 초기농도는 0.1 μ Ci/ml이었다. 산청과 은양의 점토는 흡착능력이 우수하였으나 무안의 점토는 현저하게 낮았다. 이것은 무안점토에 다량 존재하는 석영성분때문인 것으로 생각되었다.

이상의 흡착특성을 Freundlich형의 형태로 다음과 같이 표시할 수 있었다.

$$C_R = 18.0 C_A^{0.74} : \text{Cs-137}$$

$$C_R = 0.84 C_A^{0.45} : \text{Sr-90}$$

이 관계식을 BOX모델에 적용하여 점토층내에서의 핵종이동을 모사한 결과 국산점토가 처분장의 충전제로서 효과적임을 확인하였다.

1. Introduction

The long term isolation of nuclear wastes in the disposal site can be performed by introducing the multiple-barrier concept. The clay layer is important as a natural barrier among the numerous design alternatives of the disposal site, and the trench type relying on the clay barrier has been dominant for the low and medium level radioactive waste disposal since 1950s, especially in the United States. For the ten disposal sites in U.S., it has been found out that the ground water has contacted with buried wastes apparently and some portion of those has migrated considerable distances from the burial trenches.¹⁾ Many analysis on well water and core migrated at least a few meters. It has been also reported that ruthenium-106 had almost the same velocity with tritium.²⁾

In view of the long term safety analysis of the disposal site, the above mentioned experiences compelled us to perform the detailed studies to meet the domestic situations though various studies have been carried out in other countries.³⁾

This study, as a fundamental step, includes the adsorption characteristics of domestic clay sampled from Sanchong Mooan and Onyang for Cs-137 and Sr-90, and prediction of the nuclide migration behavior through clay layer. For the adsorption properties, adsorption capacities of the clays were measured in relation to solution concentration, and then, adsorption isotherms of Freundlich type were correlated with the obtained values. On the other hand, the composition of clay samples were determined by the X-ray diffraction.

To predict the nuclide migration behavior with respect to the clay layer distance and elapsed time, numerical calculation was carried out by introducing BOX model combined with

the equilibrium relationships.

2. Theory

The main compositions of clay are kaolinite and montmollilonite which have the ability to accept ions from the aqueous phase largely by sorption mechanism. The sorption has been known to take place at the exposed crystal faces physically and at the -Si-OH group replacing H^+ from it.⁴⁾

The distribution of nuclide between clay and liquid can be expressed in terms of the partition coefficient (K_{sorp}). Taking I_0 as count rate of the initial groundwater and I_t as that of contacting time t , K_{sorp} is given by,

$$K_{sorp} = \frac{\text{nuclide concentration on clay}}{\text{nuclide concentration in water}} = \frac{I_0 - I_t \exp(\lambda t)}{I_t \exp(\lambda t)} \times \frac{\alpha}{\beta} \quad (1)$$

where, λ : decay constant for the nuclide involved

α : total water content, $\alpha = V + M\phi$

β : total dry clay content, $\beta = M(1 - \phi)$

ϕ : clay porosity

M : clay weight

V : aqueous phase volume

On the other hand, as the adsorption equilibrium is dependent on the concentration of the aqueous phase, it is useful to express the equilibrium relationship in the Freundlich type isotherm.

$$C_R = KC_A^a \quad (2)$$

where, C_R : clay phase nuclide concentration

C_A : aqueous phase nuclide concentration

K, a : constant and exponent

For the simulation of radionuclide movement through clay layer, the following assumptions were made;

—The composition of the clay and groundwater flow rate are uniform throughout the clay layer.

—The concentration of radionuclide released from radwaste package is constant.

—The underground water movement is so slow that equilibrium is maintained between the aqueous phase and the clay at every step of the sub-layer(box).

With the isotherm equation and the assumptions the BOX model⁽³⁾ can be introduced for the simulation. For this the clay layer is divided in N number of sub-layers every one of which is called box. Taking the cross-section area of flow path corresponding to a unit box volume, the aqueous nuclide input to box at time t is given by ${}_{n-1}^{t-1}C_A$, while the residual clay phase content is ${}_{n-1}^{t-1}C_R(1-\phi)$. Therefore the total nuclide content in box n at time t is

$${}_n^tT = {}_{n-1}^{t-1}C_A\phi L + {}_{n-1}^{t-1}C_R(1-\phi)\rho L \quad (3)$$

where, T : total concentration of radionuclides in a box ($\mu\text{Ci}/\text{cm}^3$)

L : box length (cm)

ϕ : pore volume fraction of the clay

ρ : clay density (g/cm^3)

Equilibrium condition at box n gives,

$${}_n^tT = {}_n^tC_A\phi L + K({}_n^tC_A)^a(1-\phi)\rho L \quad (4)$$

where, K and a denotes equilibrium constant and exponent respectively for the Freundlich type isotherm equation. And, the initial and boundary conditions are

$${}_0^tC_A = \text{const} \quad (5)$$

$${}_n^0C_A = {}_n^0C_R = {}_n^0T = 0 \quad (6)$$

In practical conditions the aqueous phase concentration is very low in comparison with the clay phase concentration, that is,

$${}_n^tC_R \gg {}_n^tC_A \quad (7)$$

Now, the numerical iteration can be carried out with the equations simplified from the equations (3) and (4);

$${}_n^tC_R = {}_n^tT/\rho(1-\phi)L \quad (8)$$

$${}_n^tC_A = ({}_n^tC_R/K)^{1/a} \quad (9)$$

These values obtained for the n -th box at t -th time step are served as initial concentrations

for the next step.

3. Experimental

Clay samples from Sanchong, Onyang and Mooan were dried in the oven at 100°C , then the samples were ground and sieved to get uniform particle size of about 50 mesh. Then 1 μCi of radioactive solution was put in a batch type mixer together with a clay sample of precisely determined weight. The mixture was stirred 4 hours in a day and kept contacting for one week. Periodically, small portions of aliquot were taken and centrifuged followed by the measurement of radioactivity for the liquid phase. Everline-MCA and GM counter were used for activity measurement of Sr-90 and Cs-137, respectively. The procedure was repeated with different conditions of liquid concentration and different clay sample weight.

On the other hand, composition of the clay samples was analyzed by the X-ray diffraction method.

4. Result and Discussion

A. X-ray diffraction analysis

The X-ray diffraction spectrum⁽³⁾ of clays sampled at Sanchong, Onyang, and Mooan are shown in Fig. 1, where chlorite and kaolinite are main composition in Sanchong and Onyang clay while considerable amount of quartz is found in Mooan clay. For this measurement, bentonite and activated clay were used as reference materials.

B. Sorption coefficient

The sorption capacities obtained, under various conditions are shown in Fig. 2 and 3. The sorption equilibrium could be obtained in 4 to 6 days. The value of K_{sorp} for Sanchong clay with Sr-90 was about 14000ml/g and that of Cs-137 was about 17000ml/g. In this study the

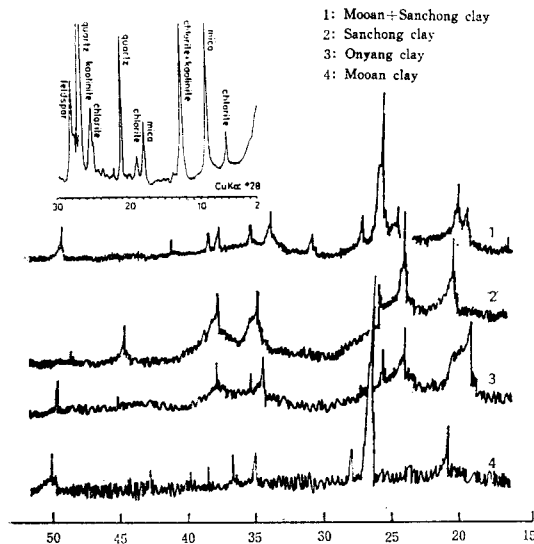


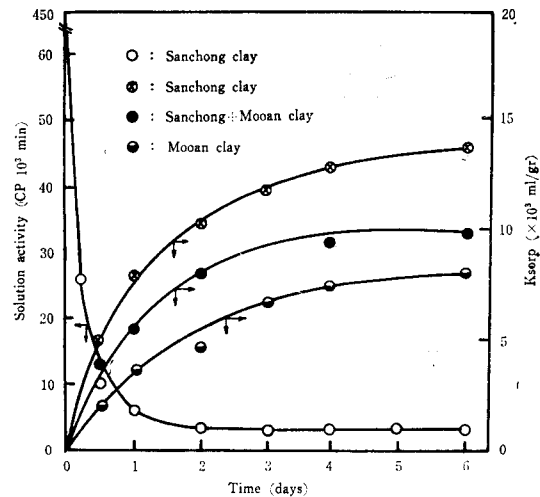
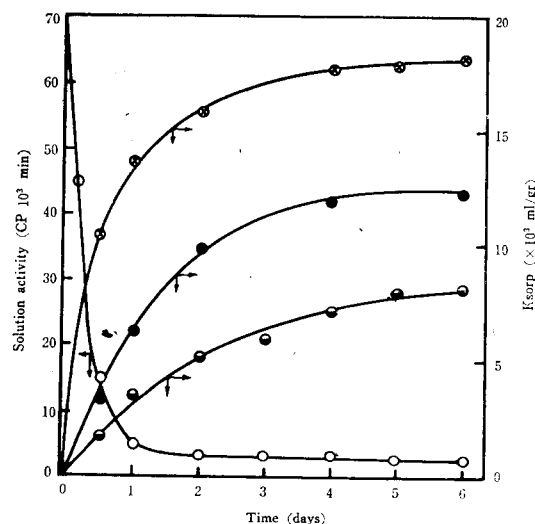
Fig. 1. X-ray Diffraction Spectrum of Clays.

Onyang clay showed almost the same sorption capacity, so comparison was made for the Sanchong clay with the Mooan clay. The Mooan clay showed very poor sorption capacity, and its K_{sorp} were about 8000ml/g for both of the nuclides. As could be expected, the mixture of Sanchong and Mooan clay (1:1) had the intermediate value of K_{sorp} . It could be explained that the quartz found in Moonan clay had poor capacity of sorption and it decreased the total capacity of the clay.

On the other hand, the values were reported to be 320ml/g for granite with Cs-137, and 2000 on Argillite.⁽⁵⁾ And, other several reports indicated much lower values for hard rock or soil.^(6,7,8)

In considering the values previously obtained in other countries, the K_{sorp} determined in this work for Sanchong and Onyang clay presented promising quality as a backfill material for the domestic radwaste disposal repository.

Now for the simulation of the underground movement the values of equilibrium sorption concentration of clay phase were plotted in log-log chart with respect to those of liquid phase

Fig. 2. Sorption of 0.1 μ Ci/ml Sr-90 on 1 gr ClaysFig. 3. Sorption of 0.1 μ Ci/ml Cs-137 on 1 gr Clays
○ : Sanchong Clay ⊗ : Sanchong Clay
· : Mooan Clay ● : Sanchong+Mooan Clay

as depicted in Fig. 4. The Freundlich type isotherm relationships were obtained;

$$C_R = 18C_A^{0.74} \text{ for Cs-137} \quad (10)$$

$$C_R = 0.84C_A^{0.45} \text{ for Sr-90} \quad (11)$$

C. Simulation of underground movement

The simulation was made by using the BOX model with the equations 10 and 11 for different box length and movement time. Fig. 5 shows that the concentration of Cs-137 is below 2×10^{-5} μ Ci/ml after 20,000 days at the distance

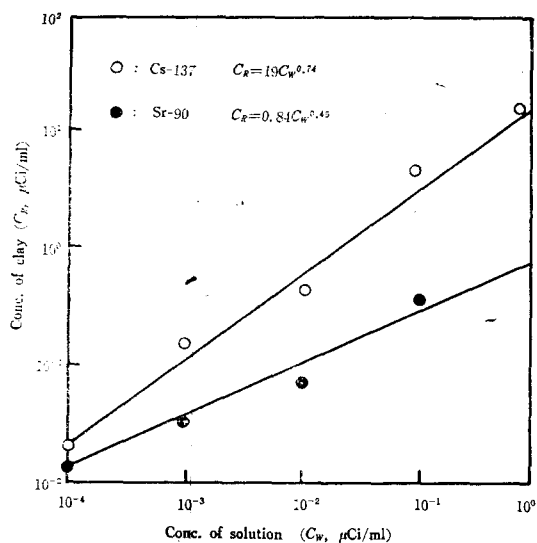


Fig. 4. Equilibrium Isotherm of Cs-137 and Sr-90

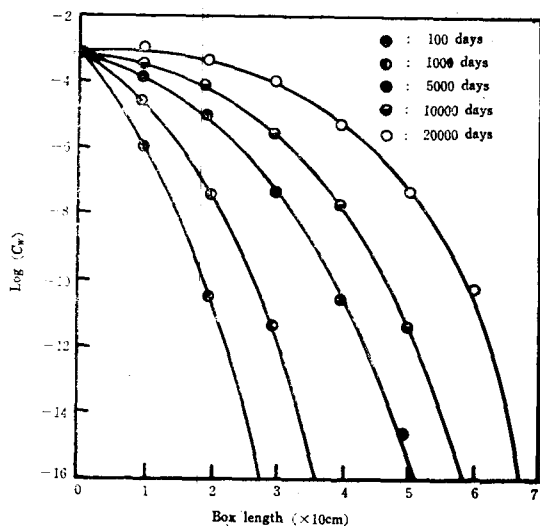


Fig. 5. Theoretical Estimation of Concentration Profile of Cs-137 in Clay Layer
Cs conc. = $1 \times 10^{-3} \mu\text{Ci/ml}$, Clay Density = 1.85
Box Length = 10cm, Clay Porosity = 0.12
Water Velocity = 1.0cm/day

of 40cm of clay barrier for the constant leakage of $1 \times 10^{-3} \mu\text{Ci/ml}$, while the concentration of Sr-90 is below $3 \times 10^{-7} \mu\text{Ci/ml}$ after 10,000 days at the distance of 50cm (see Fig. 6). The concentration criteria mentioned here are the maximum permissible concentrations of drinking water for the public. On the other hand, a plot was made in Fig. 7. to show how the leakage

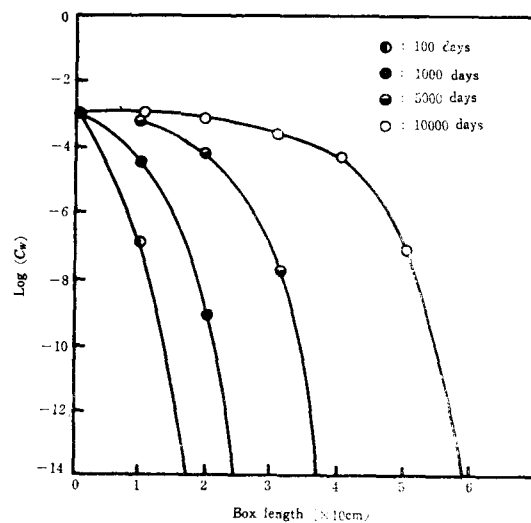


Fig. 6. Theoretical Estimation of Concentration Profile of Sr-90 in Clay Layer
Sr conc. = $1 \times 10^{-3} \mu\text{Ci/ml}$, Clay Density = 1.85
Box Length = 10cm, Clay Porosity = 0.12
Water Velocity = 1.0cm/day

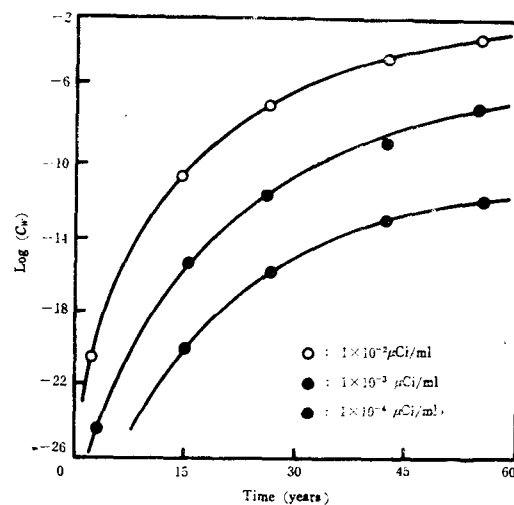


Fig. 7. Variation of Cs-137 Concentration with Time (Clay Length = 50cm)

concentration affects the diffusion rate along with the duration. For the 50cm of the domestic clay and 60 years of duration, the activity of Cs-137 reaches at more than $1 \times 10^{-4} \mu\text{Ci/ml}$ with the leakage concentration of $1 \times 10^{-2} \mu\text{Ci/ml}$, while the activity comes to less than $1 \times 10^{-12} \mu\text{Ci/ml}$ with that of $10^{-4} \mu\text{Ci/ml}$. In practice,

the leakage concentration expected at the disposal site is around $1 \times 10^{-5} \mu\text{Ci/ml}$ at least and the 50cm of clay barrier, for example, is enough to retard the release of radionuclides of low and medium level radwaste for a few hundred years. For the more, further retardation⁽⁹⁾ is expected by surrounding soil and concrete trench when engineered barriers are introduced. This research work is expected to be extended to describe the exact phenomena of release pattern of the radionuclide and multidimensional dynamic behavior^(10,11) of sorption and desorption in the ground.

5. Conclusion

The study on the sorption characteristics and underground movement for the domestic clay sampled at Sanchong, Mooan and Onyang with Cs-137 and Sr-90 showed that:

1) K_{sorp}, representing the sorption capacity, were ranged 8,000 to 17,000ml/g and 10,000 to 15,000ml/g, respectively, for Cs-137 and Sr-90 for the 0.1 $\mu\text{Ci/ml}$ of initial concentration. The clays of Sanchong and Onyang showed higher values of K_{sorp} than that of Mooan. This could be explained by poor capacity of

quartz found in Mooan clay.

2) For the quantitative analysis, Freundlich type sorption equations were established:

$$C_R = 18 C_A^{0.74} \text{ for Cs-137}$$

$$C_R = 0.84 C_A^{0.45} \text{ for Sr-90}$$

3) Simulation of the radionuclide underground movement, introducing BOX model, showed that domestic clays could be effective to meet safety criteria for the low and medium level radwaste disposal though further research works are required for the more reliable quantitative conclusion.

References

1. C.A. Heath, IAEA-SM-243/77 (1980).
2. D.G. Doles, *Science*, **215**, 1235 (1982).
3. I.G. Mckinley and J.M. West, ENPU 81-6(1981).
4. "Radioactive Waste Disposal into the Ground", IAEA-Safety Series No. 15 (1965).
5. I. Nerethieks, IAEA-SM-257/19 (1980).
6. D.B. Stewart, et al, IAEA-SM-248/97 (1980).
7. K. Schwarzer, *Nucl. Tech.*, **60**, 97 (1983).
8. I.G. Mckinley and J.M. West, ENPU 81-14(1981).
9. I. Nerethieks, IAEA-SM-243/108 (1980).
10. J. Rochon, D. Rancon and J.P. Gourmel, IAEA-SM-243/155 (1980).
11. J.F. Washburn, et al., PNL-3179 (1980).