

A NEW SAFETY ASSESSMENT MODEL FOR SHALLOW LAND BURIAL OF LLW
BASED ON MULTICOMPONENT SORPTION THEORY

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ABSTRACT

A new model on the radionuclide migration in underground environment is developed based on "multicomponent sorption theory". The model is capable of predicting the behaviors of the coexisting materials in soil-groundwater system as "multicomponent sorption phenomena" and also predicting the radionuclide migration affected by the changes of concentrations of coexisting materials. The model is not a "statistical model" but a "chemical model" based on the "ion exchange theory" and "adsorption theory". Additionally, the model is a "kinetic model" capable of estimating the effect of "rate of sorption" on the radionuclide migration. The validity of the model was checked by the results of column experiments for sorption. Finally, sample calculations on the radionuclide migration in reference shallow land burial site were carried out for demonstration.

INTRODUCTION

Development of a model on the radionuclide migration in underground environment is indispensable to the safety assessment of shallow land burial of low level radioactive waste. One of the key points for the modeling of the behavior of radionuclide in underground environment is how to reflect the effects of coexisting materials, such as ions and inorganic-organic compounds in soil-groundwater system. Formerly proposed models are, in general, based on the concept of "retardation factor" derived from "distribution coefficient (Kd)" measured by laboratory experiments.

In regard to these models, the effects of coexisting materials are taken into account by experimentally measuring the smallest Kd values, that is the most conservative values, in radionuclide-soil-groundwater system by changing the concentrations of coexisting materials. The way of reflecting the effects of coexisting materials, stated above, has two limitations as follows:

1. Not able to predict the changes of concentrations of coexisting materials in groundwater, consequently not able to estimate the "concentration phenomena" caused by the mechanism of "multicomponent sorption".
2. Requires considerable number of sorption experiments to obtain Kd values corresponding to the combination conditions for concentrations and compositions of coexisting materials.

The limitations mentioned above give us no problem as far as we consider the radionuclide migration in natural, that is chemically undisturbed, soil-groundwater system. However, some problems arise when we try to estimate following effects:

1. The effect of additional coexisting materials newly released from "engineered barrier",

such as cemented waste and concrete pit, in shallow land burial system.

2. The effect of "chemical barrier", such as soil improving agent and nuclide precipitating agent, scattered into underground to lower the mobility of radionuclide.

From the viewpoint described above, newly proposed radionuclide migration model has some advantages since the model is based on "multicomponent sorption theory".

Namely, the model is:

1. Able to simulate the behavior of coexisting materials in soil-groundwater system as the phenomena of "multicomponent sorption". Hence, also able to predict the "concentration phenomena".
2. Able to simulate the phenomena of radionuclide migration affected by the changes of concentration affected by the changes of concentrations of coexisting materials.
3. Able to predict the "multicomponent sorption phenomena" by utilizing only the data obtained from sorption experiments for single component.

In following sections, the validity of the proposed model was checked by applying it to the results of sorption column experiments for multicomponent system, and sample calculations on the radionuclide migration in reference shallow land burial site were carried out by the proposed model to demonstrate the significance of the model.

EXPERIMENTS ON MULTICOMPONENT SORPTION PHENOMENA

Column experiments were undertaken, in order to examine the effect of coexisting materials, such as Ca and Mg, on the radionuclide migration, as follows.

Certain amount of sand sampled from Osarizawa mine located at Akita prefecture in Japan was used as "adsorbent" throughout the study. Non-radioactive stable $\text{Sr}(\text{NO}_3)_2$, and $\text{Ca}(\text{NO}_3)_2$, and $\text{Mg}(\text{NO}_3)_2$ were used as "adsorbate", and they were dissolved in deionized water to provide certain concentrations of their single binary and ternary component solutions. Concentrations of Sr, Ca, and Mg were determined by Atomic Adsorption Spectrophotometer.

The conditions of column experiments are summarized in Table I. Experimental results are indicated by solid lines in Fig. 3 to Fig. 5.

In Fig. 3 and 4, it is shown that the breaktimes of Sr are considerably reduced due to the coexistence of Ca and Mg. Furthermore, it is noticeable in Fig. 5 that concentration of Ca increases to the level 1.8 times larger than initial concentration. These "concentration phenomena" are considered to be caused by additional Ca newly leached from the sand. Such phenomena are frequently observed at any soil environment, hence, it is needed to develop a new migration model which is able to explain such "concentration phenomena". The "multicomponent sorption model" is, therefore, developed by applying "multicomponent sorption theory".

PROPOSED MODEL ON MULTICOMPONENT SORPTION PHENOMENA

In this section, "multicomponent sorption model" is derived based on "mass balance equation", "rate equation for sorption" and "equilibrium equation" as follows.

Mass Balance Equation

Considering differential element of a sorption column shown in Fig. 1, following "mass balance equation" for each sorbable or exchangeable species can be introduced.

$$\gamma A [q_i(t+\Delta t) - q_i(t)] \Delta z + \epsilon A [c_i(t+\Delta t) - c_i(t)] \Delta z = uA [C_i(z) - c_i(z+\Delta z)] \Delta t + \gamma A [q_i(t+\Delta t) - q_i(t)] \Delta t \quad \text{Eq. 1}$$

$i, j = 1, 2, \dots, \text{NC}$

In Eq. (1), the effects of radioactive decay and axial dispersion are neglected and uniform presaturation of adsorbent with i -th species is assumed.

Second term of right hand side of Eq. (1) expresses exchange of i -th with j -th species in adsorbent.

Division by Δz and Δt , and taking the limits as Δz and Δt go to zero give,

$$\gamma \frac{\partial q_i}{\partial t} + \epsilon \frac{\partial c_i}{\partial t} = u \frac{\partial c_i}{\partial z} + \gamma \sum_{j=1}^{\text{NC}} \frac{\partial q_j}{\partial z} \quad \text{Eq. 2}$$

$i, j = 1, 2, \dots, \text{NC}$

Rate Equation

"Rate of sorption" for each species is expressed by Eq. (3) which is generally used in chemical engineering field. (2) (3)

$$\gamma \frac{\partial q_i}{\partial t} = K_{Fav} (c_i - c_i^*) = \beta K_{sav} (q_i^* - q_i) \quad \text{Eq. 3}$$

K_{Fav} and βK_{sav} are "overall mass transfer coefficients", and c_i^* and q_i^* are "equilibrium amount" for c_i and q_i , respectively.

Equilibrium Equation

In case that the "ion exchange" is the dominant mechanism in sorption phenomena, Eq. (4) can be employed as equilibrium relationship. (4)

$$y_i = x_i / \sum_{j=1}^{\text{NC}} a_j^i \cdot x_j \quad \text{Eq. 4}$$

where, $y_i = q_i / \text{CEC}$ and $x_i = c_i / \sum_{j=1}^{\text{NC}} c_j$

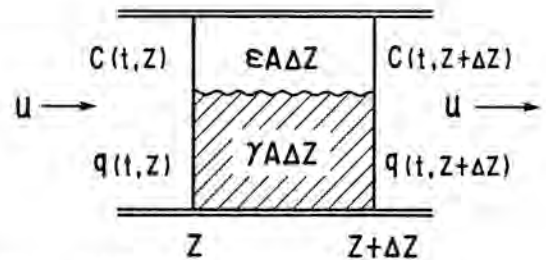


Fig. 1 Differential Element of a Sorption Column

Numerical calculations of Eq. (2), (3), and (4) can be carried out by utilizing computer. The computer program devised for the present calculation is given the code name "MUSE (Multicomponent Sorption Effect Estimation Code)", and the flow-chart of this code is shown in Fig. 2.

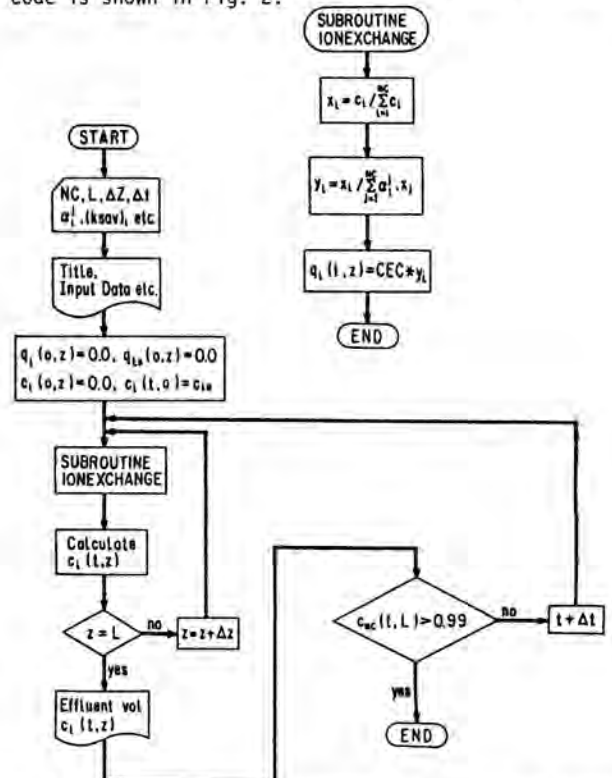


Fig. 2 Flowchart for Multicomponent Breakthrough Curve Calculation

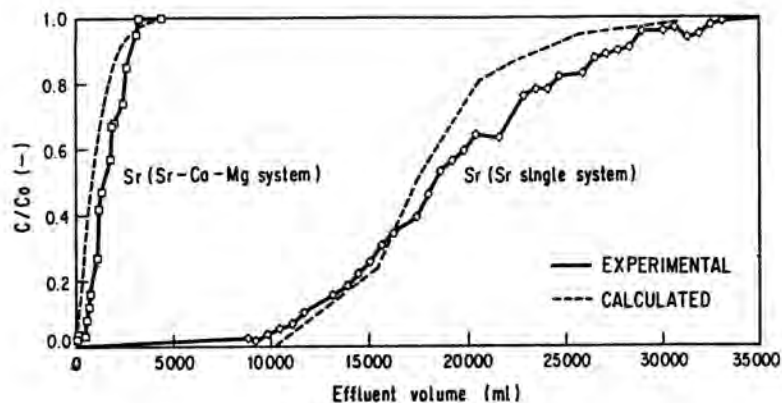


Fig. 3 Break-through curves of Sr

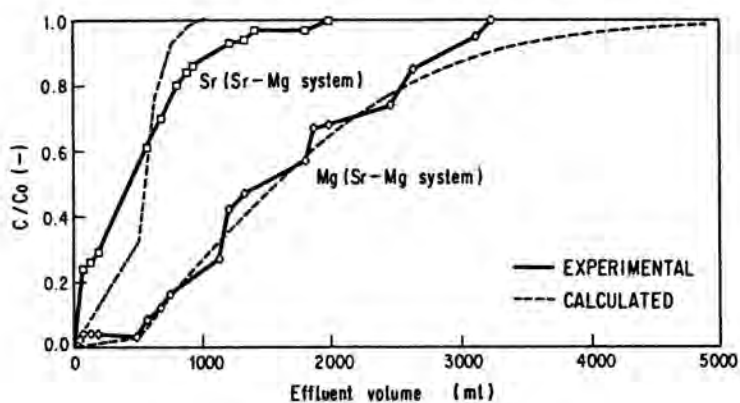


Fig. 4 Break-through curves of Mg-Sr system

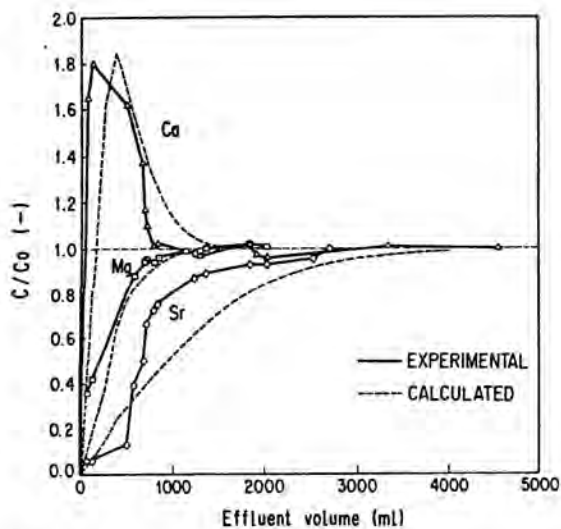


Fig. 5 Break-through curves of Sr-Ca-Mg system

Table I Conditions of Column Experiment

Sand	Particle Size	0.35 ~ 0.84 mm
	Cation Exchange Capacity	CEC = 3 meq/100g
Adsorbate	Concentration of Ca^{2+}	$C_1 (l.o) = 10\text{ppm}$
	• of Mg^{2+}	$C_2 (l.o) = 10\text{ppm}$
	• of Sr^{2+}	$C_3 (l.o) = 1\text{ppm}$
	Flow Rate	$U = 300\text{ cm/day}$
Column	"Length"	$Z = 4.5\text{ cm}$
	Cross Section Area	$A = 2.0\text{ cm}^2$
	Packed Density	$\bar{\gamma} = 1.48\text{ g/cm}^3$
	Void Fraction	$\epsilon = 0.4$

COMPARISON OF MODEL PREDICTION AND EXPERIMENTAL RESULTS

To predict the experimental results by proposed model, parameters needed for calculation, such as CEC, $(K_{sav})_i$ and (α_{ij}) , are evaluated from experimentally measured shapes of breakthrough curves indicated by solid lines in Fig. 3 and Fig. 4. As a result, 3.7 meq/100g for the value of CEC, 0.28 for (α_{SrMg}) and 10 for $(\beta K_{sav})_i$ (for all i) are determined.

However, it is difficult to determine the "separation factor" (α_{CaMg}) under the conditions of these experiments, hence, we adopt 1.0 for the value of (α_{CaMg}) . It is one of the advantages of proposed model that these parameters can be determined for each species independently.

Calculated breakthrough curves are indicated by dotted lines in Fig. 3 to Fig. 5. In Fig. 3 and Fig. 4, good agreement is observed between calculated and experimental breakthrough curves. Hence, it can be said that proposed model is able to estimate the effect of coexisting ionic materials, i.e. Ca and Mg, on the migration of "ionic state" radionuclide, for instance Sr. Calculated results for the breakthrough curves of Sr-Ca-Mg system are indicated by dotted lines in Fig. 5. In the figure, it should be noted that "concentration phenomena", in other words "overshoot phenomena", for Ca are well estimated by proposed model.

Thus it is recognized that proposed model is capable of explaining sufficiently the experimental results by using only the parameters determined from the experimental data for single species, that is, the model is capable of estimating the behaviors of radionuclides as well as coexisting materials in radionuclide-soil-groundwater system in which ion exchange is the dominant mechanism for sorption.

APPLICATION TO THE SAFETY ASSESSMENT CALCULATION FOR REFERENCE SHALLOW LAND BURIAL SITE

A new radionuclide migration model based on the "multicomponent sorption model" proposed in this study is expected to be a good support for the safety assessment of shallow land burial site by assessing the effects of coexisting materials released from engineered barrier system on the behavior of radionuclides. From above viewpoint, sample calculations for safety assessment were carried out by the "multicomponent sorption model" to demonstrate the effect of coexisting materials originated from engineered barrier on the migration of radionuclide in soil environment. For these sample calculations, two cases of "site condition and release accident" were assumed as follows:

Case 1

The groundwater in shallow land burial site contains Ca of 10 ppm and Mg of 5 ppm as "natural background concentration" in equilibrated soil-groundwater system. In this site, leakage of Sr from waste to groundwater takes place for 2 years, and the region contaminated by Sr appears at the disposal site and migrates downstream.

The initial concentration level of Sr in groundwater at disposal point caused by the leakage from waste is assumed to be 10^{-4} ppm. 30 m/day, is adopted as the value of the velocity of groundwater considering the convenience of the sample calculations.

Case 2

Addition to case 1, release of Ca and Mg into groundwater from engineered barrier system, i.e. cemented waste and concrete pit, takes place for 2 years, simultaneously, and the chemically disturbed region, where groundwater contains Ca of 20 ppm and Mg of 10 ppm as "natural background concentration + released concentration" appears and migrates downstream. The velocity of groundwater is identical to that in case 1.

Sample calculations were undertaken by MUSE code to simulate the migration of radionuclide and co-existing materials due to groundwater flow in two cases described above. Calculated results are shown in Fig. 6-(a) and (b). Figure 6-(a) indicates the spatial changes of the concentrations of Sr, Ca and Mg in groundwater 1.2 years after release accident in case 1 and 2.

Figure 6-(b) expresses the spatial changes of the concentrations of Sr, Ca and Mg after 3.0 years.

Comparing case 1 with case 2 by Fig. 6-(a) and (b), following points should be noted.

1. In case that Ca and Mg are released from engineered barrier system, the concentrations of Ca and Mg in groundwater increase. As a result, the "concentration phenomena" for Ca due to the "multicomponent sorption mechanism" is observed. Consequently, the region of higher concentration for Ca, almost 40 ppm, appears in the vicinity of disposal point and migrates downstream.
2. The migration velocity of Sr is accelerated in the region of higher concentration of Ca and Mg compared with that in the region of natural background. For instance, migration velocities of the "front" of the region contaminated by Sr (the concentration of Sr in groundwater 10^{-5} ppm is adopted to represent "front") are 58 m/year in case 1 and 146 m/year in case 2, respectively. Additionally, 1.2 years after "release accident", the range of the contaminated region (represented by the value 10^{-5} ppm) expands to 119 m downstream in case 1 and 241 m in case 2, respectively.
3. In case 2, appeared region of higher concentrations of Ca and Mg migrates downstream with time, and disappears from the range of 500 m after 3.0 years. As a result, the concentrations of Ca and Mg in the range of 500 m decrease to the background level again, that is 10 ppm for Ca and 5 ppm for Mg. However, the spatial distribution of the concentrations of Sr in case 2 largely differs from that in case 1. This is the "after-effect" of release of Ca and Mg from engineered barrier system. The disparity of the location of peak between case 1 and case 2 in Fig. 6-(b) is caused by the difference of the migration velocity of Sr between them during the period in which higher concentrations of Ca and Mg appear in case 2. On the other hand, the difference of the distribution shape between case 1 and case 2 in Fig. 6-(b) implies the effect of the "rate of sorption", that is "kinetic effect".

Thus, before the safety assessment for shallow land burial, supporting assessment by the model proposed in this study is advisable to be carried out in order to estimate the effects of following phenomena.

1. In case of that coexisting materials, i.e. Ca and Mg, are released from engineered barrier system, the maximum concentrations of coexisting materials can be larger than "natural background concentration + released concentration" due to the "concentration phenomena".
2. The migration velocity of Sr leaked from waste is accelerated and the range of contaminated region is enlarged due to the higher concentrations of coexisting materials still remains.
3. The "front" of contaminated region migrates fast in case that the "rate of sorption" is small and the reaction of sorption is slow.

The "multicomponent sorption model" and calculation code "MUSE" are able to estimate the effects of the phenomena stated above. Hence, proposed model is expected to be a good "tool" for the safety assessment of shallow land burial. In near future, proposed model will be improved to be capable of simulating not only "ion exchange phenomena" but also "adsorption phenomena" and "precipitation and

dissolution phenomena", collectively. This improvement is for the purpose of estimating the effect of "chemical barrier", such as soil improving agent and nuclide precipitating agent, scattered into underground in the vicinity of disposal site to lower the mobility of radionuclide.

REFERENCES

1. H. TASAKA, T. ASANO, Y. AKIMOTO, "A Model on the Nuclide Migration in the Unsaturated Zone", Canadian Nuclear Society: Proceedings of International Conference on Radioactive Waste Management, p. 311, (1982).
2. R. E. TREYBAL, Mass-Transfer Operations, McGraw-Hill, (1980).
3. J. S. C. HSIEH, R. M. TURIAN and C. TIEN, "Multicomponent Liquid Phase Adsorption in Fixed Bed", AIChE Journal, Vol. 23, No. 3, p. 263, (1977).
4. D. TONDEUR and G. KLEIN, "Multicomponent Ion Exchange in Fixed Beds", I & EC Fundamentals, Vol. 6, No. 3, p. 351, (1967).

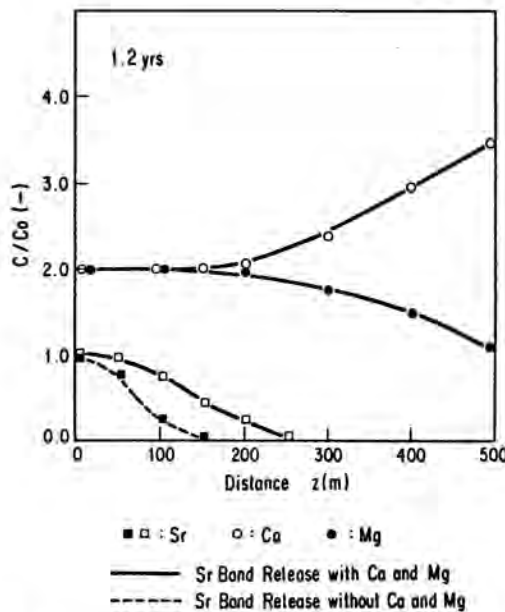


Fig. 6-(a) Sr, Ca and Mg distribution patterns in a hypothetical low-level radioactive waste disposal site

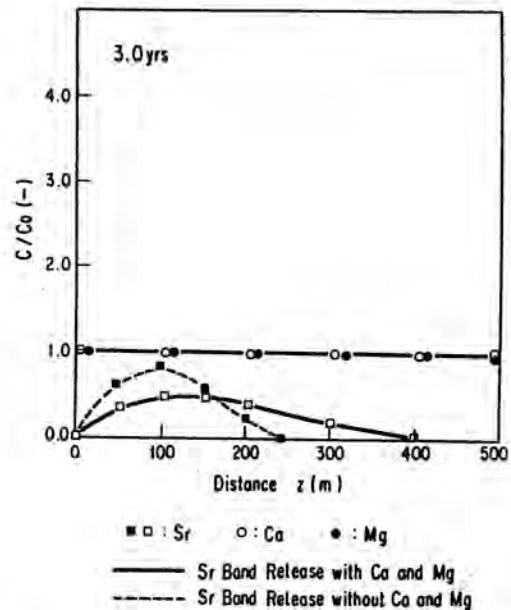


Fig. 6-(b) Sr, Ca and Mg distribution patterns in a hypothetical low-level radioactive waste disposal site