

COMPETITIVE EFFECT OF Ca AND Mg ON THE DISTRIBUTION OF TRACER RADIONUCLIDES TO THREE ION EXCHANGERS

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We theoretically considered the distribution of ⁶⁰Co and ⁸⁵Sr to ion exchangers from solution that contained Ca⁺⁺ and Mg⁺⁺ simultaneously as major cations, and derived a equation that expresses the quantity of ⁶⁰Co or ⁸⁵Sr adsorbed to an ion exchanger under the influence of these two major cations. Then we performed column experiments to determine *Kd* values of these two radionuclides in solutions containing Ca⁺⁺ and Mg⁺⁺ simultaneously to three ion exchangers, Amberlite IR-120B, Green sand and KUR sand. Experimental results of this study certifies that the equation is applicable to estimate the influence of Ca⁺⁺ and Mg⁺⁺ on adsorption of tracer ⁶⁰Co or ⁸⁵Sr.

Key Words : competitive effect, distribution coefficient, radionuclide, ion exchange resin, green sand

1. INTRODUCTION

Migration behavior of radionuclides in underground aquifers is important for the safety evaluation of underground disposal of radioactive wastes.

The distribution coefficient (*Kd*) is a parameter to present a ratio of ions adsorbed to ion exchanger from liquid phase, and commonly used in migration study of tracer radioactive cations. *Kd* is a function of ions, ion exchangers, components of ambient water, etc.^{1,2)} We examined the influence of each of several principal stable cations in ground water on *Kd* values of radionuclides by batch and column experiments. By the study, we found that *Kd* of each tracer radionuclide was presented by a simple experimental equation as a function of concentration of a stable cation if there was only one major cation in ground water³⁾. The equation is expressed as follows:

$$Kd_{AX} = Q_A / C_A = m_X \cdot (C_X)^{-n_X} \quad (1)$$

The meanings of symbols are shown in the no-

menclature preceding references. We also found that Ca⁺⁺ and Mg⁺⁺ were only two important cations in ground water that affected the *Kd* values of radionuclides⁴⁾.

In this study, we examined quantitatively the influence of coexisting Ca⁺⁺ and Mg⁺⁺ on the *Kd* values of tracer radionuclides using several parameters obtained in the previous studies. At the beginning we theoretically considered the distribution of tracer radionuclides to ion exchanger from solution that contained both Ca⁺⁺ and Mg⁺⁺ as major cations. Then we performed column experiments to determine the equilibrium constant between Ca⁺⁺ and Mg⁺⁺, and *Kd* values of radionuclides in solutions containing Ca⁺⁺ and Mg⁺⁺ simultaneously, initial concentrations of which were changed in two orders of magnitude. Cobalt-60 and Strontium-90 are considered to be important radionuclides in radioactive wastes from the viewpoint of environmental safety⁵⁾. We intended to examine the behavior of these two nuclides. Short half-life radioisotope ⁸⁵Sr was used in stead of ⁹⁰Sr in the experiments. As ion exchangers, one synthetic resin, Amberlite

Table 1 Properties of Ion Exchangers Used in the Experiment

Ion exchanger	Material	Effective grain size(mm)	Ion exchange capacity(meq/g)	Specific gravity
				(water content 40 %)
Amberlite IR-120B	Polystyrene	1.19 - 0.307	4.4	1.33
Green sand	Natural mineral	1.19 - 0.590	0.15	2.48
KUR sand	Natural mineral	0.125 - 0.250	0.031	2.47

IR-120B, and two natural ion exchangers, green sand and KUR sand, were selected in this study. Amberlite IR-120B was selected as a reference of pure synthetic ion exchanger. Green sand is a representative clay mineral commercially available. KUR sand is a sand collected from underground aquifers in a site of Research Reactor Institute, Kyoto University, Japan, and selected as a representative of natural sands. Some properties of these three ion exchangers used in this study are shown in **Table 1**.

2. THEORETICAL

Based on the results obtained by the previous studies^{3),4)}, we theoretically consider the influence of Ca^{++} and Mg^{++} ions on variation of Kd value of coexisting tracer radionuclides, ^{85}Sr and ^{60}Co . We assume the equilibrium state of two major stable cations, Ca^{++} and Mg^{++} , and tracer radioactive cations between solution and a solid phase ion exchanger. We introduce the following conditions:

- (1) The number of adsorption sites on ion exchanger does not fluctuate with changes of composition and ion concentration in solution, therefore

$$Q_X = Q_Y = Q \quad (2)$$

$$Q'_X + Q'_Y = Q \quad (3)$$

The adequacy of equation (2) was experimentally certified by our previous study⁴⁾.

- (2) When two cations X and Y are in equilibrium between a solution and an ion exchanger, the coefficient K_{XY} defined by equation (4) is constant.

$$Q'_X / C_X = K_{XY} \cdot Q'_Y / C_Y \quad (4)$$

This fact was well known if X and Y are equivalent and homologous like Ca^{++} and Mg^{++} .

Under the above conditions, we consider the three component system, two major cations, X and Y , and one tracer radionuclide A . We express their concentrations in solution as C'_X , C'_Y and C_A . Then the number of adsorption sites on an ion exchanger occupied by cation X and Y are nearly equal to Q'_X and Q'_Y , respectively, because C_A in mol concentration is very low and ignorable. We assume that X and Y are equilibrated between an ion exchanger and solution, and that a tracer radionuclide is added in solution. The number of adsorption sites displaced by radionuclide A from major cation X is notated Q'_{AX} , and expressed as follows:

$$Q'_{AX} = Q_{A(X)} \cdot (Q'_X/Q) = m_X \cdot (C_{(X)})^{-n_X} \cdot (Q'_X/Q) \cdot C_A \quad (5)$$

$$C_{(X)} = C'_X + K_{XY}^{-1} \cdot C'_Y \quad (6)$$

$Q_{A(X)}$ is the number of adsorption sites in unit weight of ion exchanger when one major cation X exists in $C_{(X)}$ in solution. Parenthesis in $C_{(X)}$ means that concentrations of ions X and Y are converted to the concentration of a single ion X by equation (6). Equation (5) shows that the influence of one major cation X in $C_{(X)}$ is the same with that of two major cations, X in C'_X and Y in C'_Y . In the same way,

$$Q'_{AY} = Q_{A(Y)} \cdot (Q'_Y/Q) = m_Y \cdot (C_{(Y)})^{-n_Y} \cdot (Q'_Y/Q) \cdot C_A \quad (7)$$

$$C_{(Y)} = C'_Y + K_{XY} \cdot C'_X \quad (8)$$

Therefore the total number of adsorption sites occupied by A is,

$$Q_A = Q'_{AX} + Q'_{AY} \quad (9)$$

$$= m_X (C_{(X)})^{-n_X} (Q'_X/Q) \cdot C_A + m_Y (C_{(Y)})^{-n_Y} (Q'_Y/Q) \cdot C_A$$

$$= \{ m_X (C_{(X)})^{-n_X} Q'_X + m_Y (C_{(Y)})^{-n_Y} Q'_Y \} \cdot (C_A/Q)$$

$$= \{ m_X (C_{(X)})^{-n_X} Q'_X + m_Y (C_{(X)})^{-n_Y} K_{XY}^{-n_Y} Q'_Y \} \cdot (C_A/Q) \quad (10)$$

The objective of this study is to evaluate the influence of Ca^{++} and Mg^{++} on the adsorption of tracer

Table 2 Values of Coefficients m_X, m_Y, n_X, n_Y

Radio-nuclide	Amberlite IR-120		Green sand		KUR sand		Amberlite IR-120		Green sand		KUR sand	
	m_X	n_X	m_X	n_X	m_X	n_X	m_Y	n_Y	m_Y	n_Y	m_Y	n_Y
^{60}Co	400	0.65	130	0.38	84	0.67	510	0.65	180	0.36	120	0.65
^{85}Sr	210	0.39	68	0.31	38	0.43	290	0.38	90	0.30	51	0.44

rem: Suffix X and Y present Ca^{++} and Mg^{++} , respectively.

m_X, m_Y, n_X and n_Y are coefficients in equation (1) when C_X and Kd_{AX} are presented in 10^{-3} meq/L and mL/g, respectively.

radionuclides, therefore we assign hereafter X in equation (10) to Ca^{++} and Y to Mg^{++} . As mentioned in Introduction, ^{60}Co and ^{85}Sr are selected as radiologically important tracer radionuclides. Values of coefficients m_X, m_Y, n_X and n_Y in equation (1) were determined in previous papers^(1,2), and are shown in Table 2. As presented in Table 2, the relation $n_X \doteq n_Y$ is applicable, and equation (10) becomes,

$$Q_A = \{m_X Q'_X + m_Y K_{XY}^{-n_Y} Q'_Y\} (C_A/Q) \cdot (C_\infty)^{-n_X} \quad (11)$$

therefore, we obtain

$$Kd_{AXY} = Q_A/C_A \\ = m_X \cdot \{Q'_X + (m_Y/m_X) \cdot K_{XY}^{-n_X} Q'_Y\} (C_\infty)^{-n_X} Q \quad (12)$$

We can calculate the distribution coefficient of radionuclide A under the influence of major two cations Ca^{++} and Mg^{++} by equation (12) and parameters m_X, m_Y, n_X and K_{XY} .

3. EXPERIMENTS AND RESULTS

We performed column experiments to determine the coefficient K_{XY} shown in equation (4) and distribution coefficients of ^{60}Co and ^{85}Sr between ion exchangers and Ca-Mg mixture solutions when concentrations of Ca^{++} and Mg^{++} varied.

(1) Determination of the equilibrium constants between Ca^{++} and Mg^{++}

The equilibrium constants between Ca^{++} and Mg^{++} for three ion exchangers, Amberlite IR-120B, green sand and KUR sand, were determined in this study. Values of cation exchange capacity of ion

exchangers were 4.4, 0.15 and 0.031 meq/g, respectively. All experiments were performed in room temperature, 20-25 °C. A procedure of experiments was as follows:

Six $\text{Ca}^{++}\text{-Mg}^{++}$ mixture solutions were prepared using their nitrates and adjusted so as to be 10^{-2} and 2×10^{-3} meq/mL of total cation concentration. Concentrations of Ca^{++} and Mg^{++} were determined by EDTA method⁷⁾. The concentration ratios of Ca^{++} to Mg^{++} were adjusted to about 1:1, 3:1 and 1:3. Five grams of each ion exchanger was packed into a separate column, and each mixture

was passed through these columns at 100 mL/hr. When concentrations of Ca^{++} and Mg^{++} at an outlet of each column coincided with those at an inlet, concentrations of Ca^{++} and Mg^{++} were considered to be in equilibrium between mixture solutions and ion exchangers. Quantities of adsorbed Ca^{++} and Mg^{++} were determined by desorbing cations from ion exchangers using 2N-NaCl solution.

The coefficient K_{XY} were calculated using equation (4) and are shown in Table 3. Values of the coefficient varied only slightly with concentrations of Ca^{++} and Mg^{++} in solution for each ion exchanger, and average values are 1.9, 2.0 and 1.7 for Amberlite IR-120B, green sand and KUR sand, respectively.

(2) Determination of the distribution coefficients of ^{60}Co and ^{85}Sr

The distribution coefficients of ^{60}Co and ^{85}Sr between $\text{Ca}^{++}\text{-Mg}^{++}$ mixture solution and three ion exchanges were determined as follows. The procedure of each run was repeated two times, and the mean value of two runs was used for discussion.

Calcium nitrate and magnesium nitrate were diluted by deionized water to prepare $\text{Ca}^{++}\text{-Mg}^{++}$ mixture solutions, whose total cation concentration ranged $5 \times 10^{-3} \sim 10^{-1}$ meq/mL. Cobalt-60 or

Table 3 Values of Equilibrium Constants between Ca^{++} and Mg^{++} for Various Combinations of Solutions and Ion Exchangers

Initial concentration in Ca^{++} - Mg^{++} mixture solution (meq/mL)		Equilibrium constants between Ca^{++} and Mg^{++} ($K_{X,Y}$)		
Ca^{++}	Mg^{++}	Amberlite IR-120B	Green sand	KUR sand
4.90×10^{-2}	4.95×10^{-2}	1.92	2.05	1.61
1.04×10^{-3}	1.08×10^{-3}	1.85	2.14	1.70
7.35×10^{-2}	2.56×10^{-2}	2.01	2.10	1.65
1.48×10^{-3}	5.35×10^{-4}	1.82	1.95	1.82
2.45×10^{-2}	7.25×10^{-2}	1.83	2.03	1.73
5.62×10^{-4}	1.45×10^{-3}	1.95	1.98	1.66
Average		1.9	2.0	1.7

Table 4 Distribution Coefficients of ^{60}Co for Three Ion Exchanges

Initial concentration		Amberlite IR-120		Green sand		KUR sand	
Ca^{++} (meq)	Mg^{++} (meq)	K_d (mL/g)	$C_{(X)}$ (meq/mL)	K_d (mL/g)	$C_{(X)}$ (meq/mL)	K_d (mL/g)	$C_{(X)}$ (meq/mL)
4.9×10^{-2}	5.5×10^{-2}	24	7.5×10^{-2}	27	7.4×10^{-2}	4.5	7.8×10^{-2}
8.1×10^{-2}	2.0×10^{-2}	20	9.2×10^{-2}	24	9.1×10^{-2}	4.2	9.3×10^{-2}
2.1×10^{-2}	7.9×10^{-2}	29	6.3×10^{-2}	28	6.1×10^{-2}	6.0	6.7×10^{-2}
1.1×10^{-2}	1.0×10^{-2}	71	1.6×10^{-2}	40	1.6×10^{-2}	14	1.7×10^{-2}
1.6×10^{-2}	4.1×10^{-3}	57	1.8×10^{-2}	42	1.8×10^{-2}	11	1.8×10^{-2}
4.2×10^{-3}	1.6×10^{-2}	70	1.3×10^{-2}	56	1.2×10^{-2}	14	1.4×10^{-2}
5.2×10^{-3}	5.1×10^{-3}	95	7.9×10^{-3}	57	7.8×10^{-3}	18	8.2×10^{-3}
8.2×10^{-3}	1.9×10^{-3}	110	9.2×10^{-3}	48	9.2×10^{-3}	16	9.3×10^{-3}
2.1×10^{-3}	8.0×10^{-3}	120	6.3×10^{-3}	64	6.1×10^{-3}	30	6.8×10^{-3}
1.2×10^{-3}	1.1×10^{-3}	300	1.8×10^{-3}	110	1.8×10^{-3}	58	1.8×10^{-3}
1.6×10^{-3}	4.2×10^{-4}	250	1.8×10^{-3}	92	1.8×10^{-3}	48	1.8×10^{-3}
3.8×10^{-4}	1.7×10^{-3}	340	1.3×10^{-3}	110	1.2×10^{-3}	73	1.4×10^{-3}
5.2×10^{-4}	5.3×10^{-4}	430	8.0×10^{-4}	150	7.9×10^{-4}	94	8.3×10^{-4}

rem: $C_{(X)}$ is defined in equation (6)

strontium-85 was added to an aliquot of each mixture so as to be about 370 Bq/mL (denominated hereafter Co solution or Sr solution).

Values of pH of the Co solutions and Sr solutions were 5.5 - 6.5. Five grams of each ion exchanger was packed into a separate column, and one of the mixture solutions was passed through this column to saturate the ion exchanger with Ca^{++} and Mg^{++} . After interstitial water in a column was washed out by deionized water, ion exchanger was taken out and dried by suction.

Three grams of each ion exchanger saturated with a Ca^{++} - Mg^{++} mixture, was taken in a beaker. Then 150 mL of Co solution or Sr solution with the same Ca^{++} and Mg^{++} concentrations to the Ca^{++} -

Mg^{++} mixture was added into this beaker. This sample was settled 24 hours with periodical agitation. After settling, the ion exchanger was filtered (TOYO Co. Ltd.: No.4). It was certified by the preliminary experiment that no adsorption of radionuclide was observed to any ion exchanger after this filtration. An aliquot of the filtrate was submitted to radioactivity measurement with NaI scintillation counter, and Ca^{++} and Mg^{++} concentrations of another aliquot were determined by EDTA method.

Concentrations of Ca^{++} and Mg^{++} did not vary during 24 hours settling. Adsorbed quantities of ^{60}Co and ^{85}Sr were determined by subtracting the concentrations after settling from initial con-

Table 5 Distribution Coefficients of ^{85}Sr for Three Ion Exchanges

Initial concentration		Amberlite IR-120		Green sand		KUR sand	
Ca^{++} (meq)	Mg^{++} (meq)	K_d (mL/g)	$C_{(x)}$ (meq/mL)	K_d (mL/g)	$C_{(x)}$ (meq/mL)	K_d (mL/g)	$C_{(x)}$ (meq/mL)
5.2×10^{-2}	5.0×10^{-2}	39	7.8×10^{-2}	27	7.7×10^{-2}	5.7	8.1×10^{-2}
8.3×10^{-2}	1.8×10^{-2}	36	9.2×10^{-2}	24	9.2×10^{-2}	5.1	9.4×10^{-2}
1.6×10^{-2}	8.8×10^{-2}	44	6.2×10^{-2}	28	6.0×10^{-2}	6.6	6.8×10^{-2}
1.0×10^{-2}	1.4×10^{-2}	65	1.7×10^{-2}	31	1.7×10^{-2}	10	1.8×10^{-2}
1.9×10^{-2}	4.8×10^{-3}	68	2.2×10^{-2}	28	2.1×10^{-2}	10	2.2×10^{-2}
4.2×10^{-3}	1.9×10^{-2}	75	1.4×10^{-2}	37	1.4×10^{-2}	14	1.5×10^{-2}
5.1×10^{-3}	5.3×10^{-3}	98	7.9×10^{-3}	33	7.8×10^{-3}	17	8.2×10^{-3}
8.5×10^{-3}	1.6×10^{-3}	79	9.3×10^{-3}	33	9.3×10^{-3}	13	9.4×10^{-3}
1.6×10^{-3}	9.2×10^{-3}	120	6.4×10^{-3}	45	6.2×10^{-3}	17	6.4×10^{-3}
1.1×10^{-3}	1.4×10^{-3}	180	1.8×10^{-3}	52	1.8×10^{-3}	28	1.9×10^{-3}
1.8×10^{-3}	5.0×10^{-4}	160	2.1×10^{-3}	55	2.1×10^{-3}	28	2.1×10^{-3}
4.5×10^{-4}	1.9×10^{-3}	170	1.5×10^{-3}	62	1.4×10^{-3}	37	1.6×10^{-3}
5.1×10^{-4}	5.0×10^{-4}	250	7.7×10^{-4}	72	7.6×10^{-4}	40	8.0×10^{-4}

* $C_{(x)}$ is defined in equation (6)

Table 6 Calculated Values of $(m_y/m_x) \cdot K_{XY}^{-n_x}$

Ion exchange	Radio-nuclide	$\frac{m_y}{m_x}$	K_{XY}	n_x	$\frac{m_y}{m_x} \cdot K_{XY}^{-n_x}$
Amberlite IR-120B	^{60}Co	1.3	1.9	0.65	0.9
	^{85}Sr	1.4		0.39	1.1
Green sand	^{60}Co	1.4	2.0	0.38	1.1
	^{85}Sr	1.3		0.31	1.0
KUR sand	^{60}Co	1.4	1.7	0.67	1.0
	^{85}Sr	1.3		0.43	1.0

rem: Suffix X and Y present Ca^{++} and Mg^{++} , respectively.
 m_x , m_y and n_x are coefficients defined in equation (1).

(12) is simplified and expressed as

$$Kd_{A,XY} = m_x \cdot (C_{(x)})^{-n_x} \quad (13)$$

Equation (13) expresses that Kd of a radionuclide in Ca^{++} - Mg^{++} mixture solution can be determined by the same equation with equation (1) that presents the relation of Kd in Ca^{++} solution. Transforming the both sides of equation (13) into logarithms, we get

$$\log(Kd_{A,XY}) = \log(m_x) - n_x \cdot \log(C_{(x)}) \quad (14)$$

centrations. Kd values of ^{60}Co and ^{85}Sr were calculated by equation (1). These values are presented in **Tables 4 and 5** with concentrations of Ca^{++} and Mg^{++} in initial Co solution or Sr solution. Values of $C_{(x)}$ in equation (6) are also shown in the tables.

4. DISCUSSION

We calculate the values of $(m_y/m_x) \cdot K_{XY}^{-n_x}$ in equation (12) for ^{60}Co and ^{85}Sr using the values of m_x , m_y and n_x in **Table 2**, and K_{XY} in **Table 3**. These calculated values are presented in **Table 6**. As shown in **Table 6**, the values of $(m_y/m_x) \cdot K_{XY}^{-n_x}$ are considered nearly unity for all cases. Using this relation and equation (3), equation

The data of $Kd_{A,XY}$ versus $C_{(x)}$ presented in **Tables 4 and 5** are plotted in log-log scale in **Figs. 1 and 2**, respectively. Linear regression lines in log-log scale for each ion exchanger are also presented in the figs. Values of $m_{(x)}$ and $n_{(x)}$ determined by linear fitting of equation (14) to the data are also shown in **Table 7** for three ion exchangers, which present almost equal values with m_x and n_x shown in **Table 2**. Linearity between $\log(Kd_{A,XY})$ and $\log(C_{(x)})$ are expressed in **Figs. 1 and 2**, and can be certified by the values of correlation coefficients in **Table 7**. From these results, we can conclude that equation (13) is applied to estimate the influence of Ca^{++} and Mg^{++} on adsorption of tracer ^{60}Co or

Table 7 Values of m_{00} and n_{00} Determined by Curve Fitting of Equation (14) to the Data Shown in Tables 4 and 5.

Ion exchanger	Radio-nuclide	m_{00}	n_{00}	Correlation coefficient
Amberlite IR-120B	^{60}Co	390	0.64	-1.0
	^{85}Sr	220	0.40	-0.99
Green sand	^{60}Co	120	0.37	-0.99
	^{85}Sr	62	0.22	-0.96
KUR sand	^{60}Co	82	0.65	-0.99
	^{85}Sr	39	0.44	-0.99

^{85}Sr .

5. CONCLUSION

In this study, we examined the influence of Ca^{++} and Mg^{++} on adsorption of ^{60}Co or ^{85}Sr when tracer ^{60}Co or ^{85}Sr existed with major Ca^{++} and Mg^{++} in solution. We theoretically derived the following equation that expresses the quantity of ^{60}Co or ^{85}Sr to an ion exchanger under the influence of major two cations.

$$Q_A = \left\{ m_x (C_{00})^{-n_x} Q'_x + m_y (C_{00})^{-n_y} K_{xy}^{-n_y} Q'_y \right\} (C_A/Q) \quad (10)$$

where

$$C_{00} = C'_x + K_{xy}^{-1} C'_y \quad (6)$$

It was pointed out in the previous papers by the authors^{3,4)} that Ca^{++} and Mg^{++} were only two important cations in underground water influencing the adsorption of radionuclides. Therefore we assigned Ca^{++} and Mg^{++} to X and Y , respectively. The relation $n_x \approx n_y$ is applicable in this case, and the above equation is revised to,

$$Q_A = \left\{ m_x Q'_x + m_y K_{xy}^{-n_y} Q'_y \right\} (C_A/Q) \cdot (C_{00})^{-n_x} \quad (11)$$

then we obtain

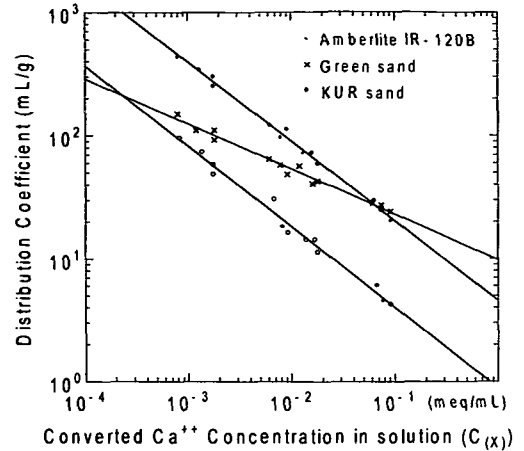


Fig.1 Distribution coefficient of ^{60}Co between three ion exchangers and Ca^{++} - Mg^{++} mixture solution. Linear regression lines are also expressed.

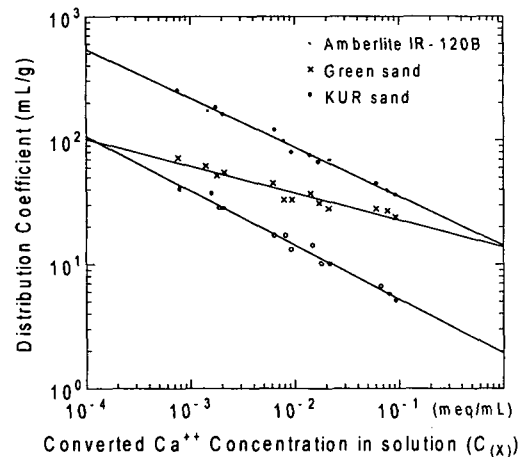


Fig.2 Distribution coefficient of ^{85}Sr between three ion exchangers and Ca^{++} - Mg^{++} mixture solution. Linear regression lines are also expressed.

$$Kd_{AXY} = Q_A/C_A$$

$$= m_x \cdot \left\{ Q'_x + (m_y/m_x) \cdot K_{xy}^{-n_x} Q'_y \right\} (C_{00})^{-n_x} / Q \quad (12)$$

On the basis of the experimental results of this study, we obtained

$$(m_Y/m_X) \cdot K_{XY}^{-n_X} \approx 1$$

Equation (12) is simplified to

$$Kd_{AXY} = m_X \cdot (C_{(X)})^{-n_X} \quad (13)$$

This equation is identical with equation (1) that expresses the relation between Kd of radionuclide A and Ca^{++} concentration in solution. In other words, the influence of both Ca^{++} and Mg^{++} can be replaced with that of Ca^{++} only by converting concentrations of Ca^{++} and Mg^{++} in solution to Ca^{++} concentration using equation (6).

Experimental results of this study certifies that equation (13) is applicable to estimate the influence of Ca^{++} and Mg^{++} on adsorption of tracer ^{60}Co or ^{85}Sr . By the result obtained in this study, we can predict the migration of tracer ^{60}Co or ^{85}Sr in natural underground aquifers more precisely than by conventional two component model.

NOMENCLATURE

- C_A : Concentration of radionuclide A in solution (Bq/mL).
- C_X, C_Y : Concentration of major stable cation X or Y in solution (meq/mL).
- C'_X, C'_Y : Concentration of stable cation X and Y in solution when two major cations X and Y exist in solution (meq/mL).
- $C'_{(X)}, C'_{(Y)}$: Converted concentration of stable cation X or Y in solution (meq/mL).
- K_{XY} : A coefficient defined by equation (4).
- Kd_{AX} : Distribution coefficient of radionuclide A when only one major cation X exists in solution (mL/g).
- Kd_{AXY} : Distribution coefficient of radionuclide A when two major cations X and Y exist in solution (mL/g).
- m_X, n_X : Parameters of equation (1) determined by a combination of radionuclide A , major cation X and an ion exchanger.
- Q, Q_X, Q_Y : The number of adsorption sites on an ion exchanger when only one cation X or Y exists in a solution.
- Q_A : The total number of adsorption sites on

an ion exchanger occupied by radionuclide A .

Q_{AX}, Q_{AY} : The number of adsorption sites on an ion exchanger occupied by radionuclide A when exists one major cation X or Y in solution, concentration of which is C_X or C_Y .

$Q_{A(X)}$: The number of adsorption sites on an ion exchanger occupied by radionuclide A when converted concentration of X in solution is $C_{(X)}$.

Q'_{AX}, Q'_{AY} : The number of adsorption sites on an ion exchanger displaced by tracer radionuclide A from X or Y , when two major cations X and Y exist in solution, concentrations of which are C_X and C_Y , respectively.

Q'_X, Q'_Y : The number of adsorption sites on an ion exchanger occupied by cation X or Y when cations X and Y exist in solution.

Subscript:

X, Y : Presents major cation.

A : Presents tracer radionuclide.

REFERENCES

- 1) M.F.Lima and B.P.Mazzilli ; Determination of the distribution coefficients for ^{134}Cs , ^{60}Co and ^{234}Th in the Pinheiros river sediment-water, *J. Radioanal. Nucl. Chem., Articles*, Vol.177, No.1, pp.139-147, 1994.
- 2) R.J.Serne; Conceptual adsorption models and open issues pertaining to performance assesment, *PNL-SA-20045*, 1991.
- 3) T.Tsutsui and K.Nishimaki ; Safety evaluation of the ground disposal of radioactive wastes(I), *Hoken Butsuri*, Vol. 10, pp.79-86, 1975 (in Japanese).
- 4) T.Tsutsui and K.Nishimaki ; Safety evaluation of the ground disposal of radioactive wastes(II), *Hoken Butsuri*, Vol.10, pp.203-210, 1975 (in Japanese).
- 5) United Nations Scientific Committee on the Effects of Atomic Radiation: *Ionizing radiation: sources and biological effects*, United Nations Publication, New York, 1982.
- 6) M. Honda; *Ion Exchange*, p26, Nanko-do, Tokyo, 1962 (in Japanese).
- 7) A.E. Greenberg et al. Ed.; *Standard Methods for the Examination of Water and Wastewater*, 18th edition, American Public Health Association, Wash., 1992

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3種類のイオン交換体への放射性核種の分配に対する Ca と Mg の影響

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放射性核種の地中処分の安全性に関して重要な核種である ^{60}Co と ^{87}Sr のイオン交換体への分配に地下水中の Ca^{++} と Mg^{++} イオンが大きな影響を与える。本研究では Ca^{++} と Mg^{++} イオンが同時に存在する環境下での放射性核種の分配を理論的に検討し、分配係数を簡単な数式で表現した。次に Amberlite IR-120B, グリーンサンド, KUR砂の3種類のイオン交換体を用いてカラム実験を行い、 Ca^{++} と Mg^{++} の平衡定数と Ca^{++} と Mg^{++} 共存下での ^{60}Co と ^{87}Sr の分配係数を求めた。実験結果と比較することにより、上記の実験式の有効性が確認された。