



#### 4.7 Experimental and Analytical Study on Removal of Strontium from Cultivated Soil

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**Abstract ;** Experimental and analytical study was done to estimate the removal of strontium from cultivated soil. The continuous batch tests were made and uneasy desorption form or immobility form was proved to exist. 2-Component Model, which considers easy desorption and uneasy desorption form fraction, was constructed and it showed good explanation of the continuous batch test results.

**Key Words ;** Strontium , Batch Test , Cultivated Soil , Sorption/Desorption , 2-Component Model

##### 1. Introduction

For a mid/long term environmental assessment of radionuclides added to cultivated soil by an accident of nuclear power plants, 1-component model, which is constructed on an assumption of distribution equilibrium between liquid and solid phase in the soil, has been used generally. On the other hand, it has been reported that an increase in frequency of water replace of the soil-water batch system led to a decrease in amount of the trace element eluted from the soil<sup>1)</sup>. It has been also observed that 2-Component Model, which was composed of fast and slow migration rates of the element, gave better explanation of monitoring data of strontium-90 in Japanese cultivated soil<sup>2)</sup>. In the present report we have studied the removal of strontium from cultivated soil experimentally and analytically.

##### 2. Experimental

In this study three soil-solution systems, that solutions were stable strontium solution, calcium solution and radioactive strontium-85 (hereafter, abbreviated Sr-85) solution, were put to the continuous batch test. The soil was sampled from the paddy field at Agricultural, Food and Environmental Science Research Center of Osaka Prefecture. The stable strontium solution was prepared from SrCl<sub>2</sub>, the calcium solution was prepared from CaCl<sub>2</sub>, and the Sr-85 was obtained in solution state with HCl. Three types of concentration were prepared for stable strontium and calcium solutions, respectively. Those were 2.4×10<sup>1</sup>, 4.8×10<sup>-1</sup>, 2.4×10<sup>-2</sup> mg/L for the stable strontium, and 3.2×10<sup>1</sup>, 6.5×10<sup>-1</sup>, 3.2×10<sup>-2</sup> mg/L for the calcium. These three types were denoted 'high(concentration)', 'middle(conc.)', and 'low(conc.)' from now. Concentration of the strontium-85 was 5.0×10<sup>4</sup> Bq/L at the start of experiments. pH values of the solutions were 5.7~6.1, provided that pH of the Sr-85 solution was controlled by (CH<sub>3</sub>)<sub>4</sub>NOH. The continuous batch test was defined in this study as follows; first, the each solutions and the soil were mixed in a sample tube, and its solid/liquid ratio was 1/10; here, for the stable strontium systems and calcium systems, the soil amount was 3g and the solution amount was 30ml, for the Sr-85 systems, the soil amount was 1.2g and the solution amount was 12ml. Then the tubes were shaken by hands once a day, and after 7 days elapsed, the tubes were centrifuged (3000rpm, 10min.) and supernatant liquids were collected for measurements. Here, the amount of collecting supernatant liquid was 5/6 of a whole liquid in one sample tube; that was 25ml for the stable strontium systems and calcium systems and 10ml for the Sr-85 systems. And after collecting, ultra pure water was added into the tubes. This replacement of each solution had been done every 7 days and the batch test was

continued for 10(for stable elements) or 14(for Sr-85) weeks. The collected liquid samples were measured by Inductive Coupled Plasma - Mass Spectrometry(ICP-MS,H.Packard;HP4500) for stable strontium,by Atomic Absorption Spectroscopy(AAS,Shimadzu;AA-646)for calcium,and by Germanium semiconductor detector for radioactive Sr-85(Ortec;GMX30190-p).

**3.Results and Analytical**

For stable strontium systems and calcium systems,we got 12 plotted curves of relationship between concentration in the liquid phase and times of the liquid replacement,those were strontium concentration in the strontium solution systems,strontium concentration in the calcium systems,calcium concentration in the strontium systems,calcium concentration in the calcium systems,and 3 types(high,middle,low) of concentration for each solution.Those plots were shown in Fig.1,here,'Sr-Ca\_H' means 'concentration of strontium in the high calcium concentration solution system'.Sr-Sr plots shows strontium concentration in

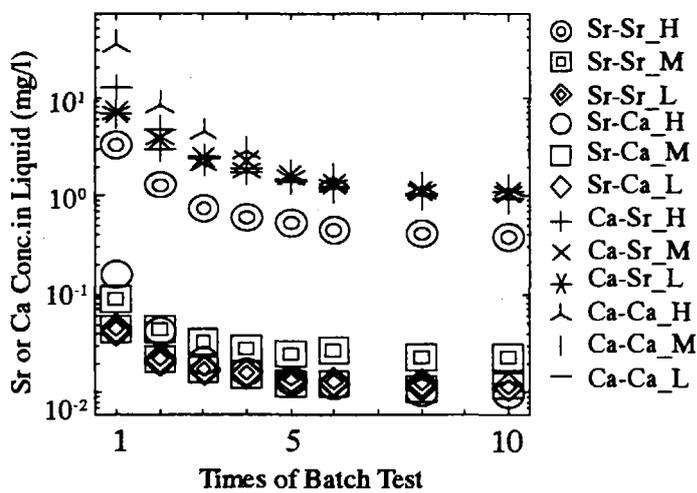


Fig.1 Results of Continuous Batch Test ; Stable Sr and Ca System  
e.g. 'Sr-Ca\_H' means 'concentration of strontium in the high calcium concentration solution system'  
Comparing with strontium concentration values in the calcium solution systems,it appears that salts concentration have little effect on the desorption of stroutium.

the liquid phase decreases gradually with increase in times of replacement. But even after 10 weeks (i.e. 10 times of replacement),strontium was detected in liquid phase,and the concentration values were in order of initial concentration of solution.That means strontium which had sorbed on the soil at the first solid-liquid contact was desorped from the soil.Comparing with strontium concentration values in the calcium solution systems,it appears that salts concentration have little effect on the desorption of stroutium.

In Fig.2,the results of mesuaring strountium-85 are plotted with times of the replacement of liquid as abscissa against Sr-85 concentration (its decay is corrected) in liquid and solid-liquid distribution ratio of Sr-85 as ordinates. In the same way as stable strontium,Sr-85 concentration in liquid phase decreases gradually with increase in times

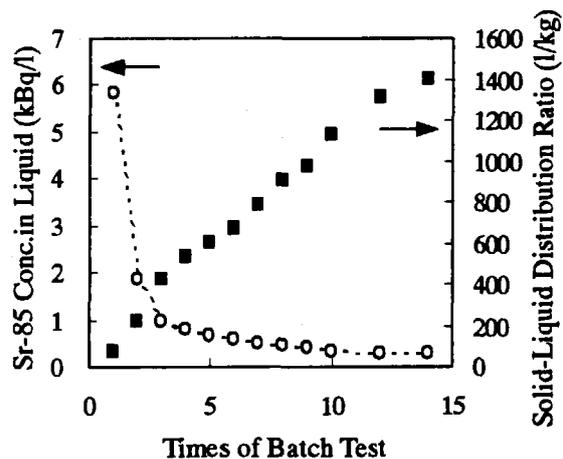


Fig.2 Results of Continuous Batch Test and Solid-Liquid Distribution Ratio; Sr-85 System

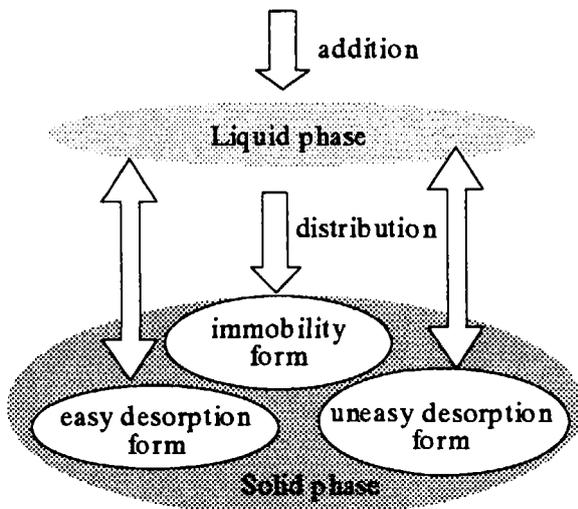


Fig.3 Conception diagram of this model

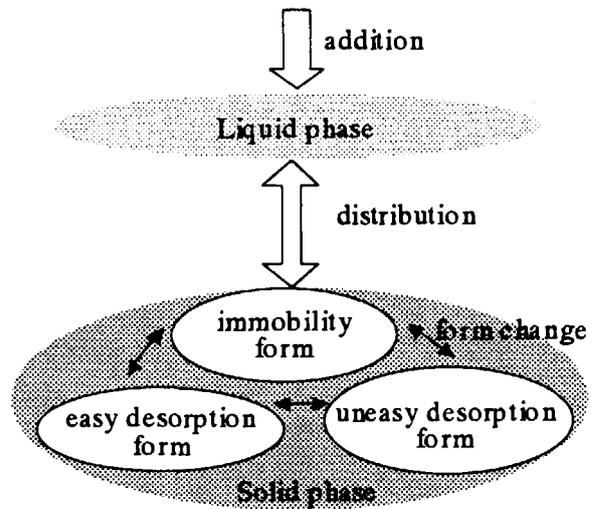


Fig.4 Conception diagram of the actual environment

of replacement. Fig.2 also shows that solid-liquid distribution ratio of Sr-85 increases, and it means that the Sr-85 rate of sorption fast on the soil increases.

A conception diagram of this model is shown in Fig.3. In the actual environment, a form of the three, those are easy desorption, uneasy desorption, and immobility form, is supposed to change to the other form (shown in Fig.4), but in this study the changes of the form are neglected. As shown in Fig.3, it is assumed that the added elements are sorbed as each form and don't change the form.

Fig.5, and Fig.6 show measurement values and the regression curves which were obtained from Eq.(1) or Eq.(2) by the least squares method.

$$C_n/C_0 = q_1(1-Rr_1)^{n-1}r_1 + q_2(1-Rr_2)^{n-1}r_2 \quad \dots\dots\dots \text{Eq(1)}$$

$$C_n/C_1 = \{p(1-Rr_1)^{n-1}r_1 + (1-p)(1-Rr_2)^{n-1}r_2\} / \{pr_1 + (1-p)r_2\} \quad \dots\dots\dots \text{Eq(2)}$$

Where,  $C_n$ ; concentration of the element in liquid at the nth replacement (Bq/l or mg/l),  $C_0$ ; initial concentration of the element in liquid (Bq/l or mg/l),  $q_1$ ; ratio of easy desorption form fraction (-),  $q_2$ ; ratio of uneasy desorption form fraction (-),  $r_1$ ; removal rate constant of easy desorption (-),  $r_2$ ; removal rate constant of uneasy desorption form fraction (-),  $R$ ; ratio of replacement liquid volume to whole liquid volume;  $R=25/30$  or  $10/12 = 5/6$ ,  $p$ ; ratio of easy desorption form fraction to whole desorption from fraction;  $p=q_1/(q_1+q_2)$

When initial amount of the element in the system

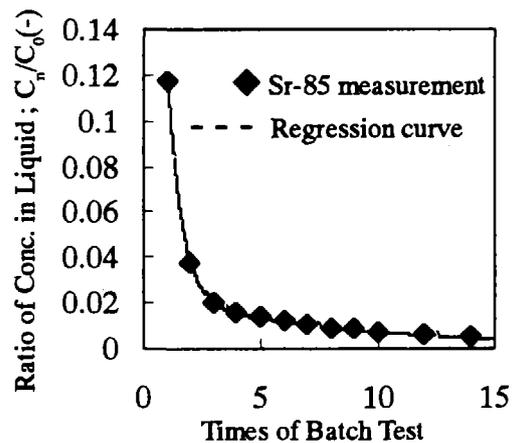


Fig.5 Regression Curve and Measurement Values of Continuous Batch Test; Sr-85 System; Regression curve is obtained from  $C_n/C_0$  model by the least squares method and Parameters are  $q_1=0.100$ ,  $q_2=0.163$ ,  $r_1=0.960$ ,  $r_2=0.127$

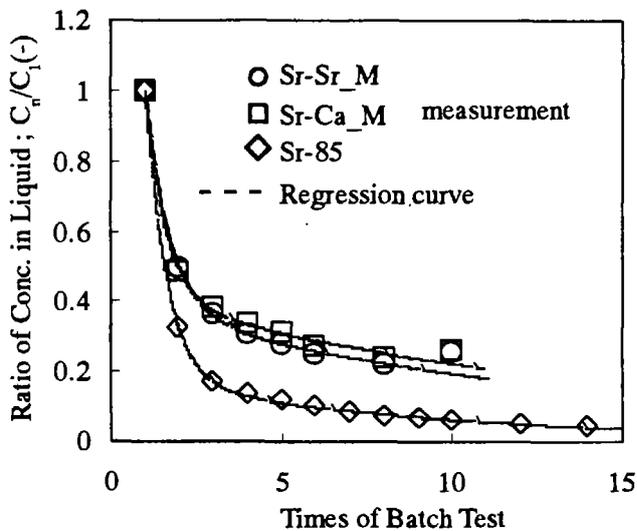


Fig.6 Regression Curve and Measurement Values of Continuous Batch Test; Sr-Sr\_M, Sr-Ca\_M and Sr-85 System; Regression curve is obtained from  $C_n/C_1$  model by the least squares method and Parameters are follow.

	p	r <sub>1</sub>	r <sub>2</sub>
Sr-85	0.383	0.955	0.121
Sr-Ca_M	0.103	0.972	0.075
Sr-Sr_M	0.133	0.901	0.081

is known, we can estimate the concentration of the element in liquid phase by using Eq(1). In this study the Sr-85 system is applicable to the case. On the other hand, when initial amount of the element in the system is unknown, Eq(1) cannot be used because the immobility form fraction cannot be known. Therefore Eq(2), in which ratio(p) is used for  $q_1$  and  $q_2$ , is used for the case of stable strontium and calcium system. The regression curves show good coincidence with measurement values in Fig.5 and Fig.6. The analytical results show that the 2-component model is useful for the estimation of distribution from soil to liquid phase of strontium.

#### 4. Summary

Continuous batch test shows increase of distribution ratio with times of water replacement. That means uneasy desorption form or immobility form exist in the system. 2-Component Model, which considers easy desorption and uneasy desorption form fraction (immobility form included, it is 3-Component), gives good explanation for the experiment

#### References

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