Migration of $^{137}\text{Cs}$ Adsorbed on Fine Soil Particles through Soil Layer

Filtration by Unsaturated Sandy Soil Layer

Hiromichi OGAWA

Department of Environmental Safety Research,
Tokai Research Establishment,
Japan Atomic Energy Research Institute*

Received October 1, 1987

The experiments on the migration of $^{137}\text{Cs}$ adsorbed on fine soil particles in unsaturated sandy soil layers were carried out. The soil layer reduced the concentration of $^{137}\text{Cs}$ in the effluent, but did not retard its migration. The concentration of $^{137}\text{Cs}$ remained in the soil layer increased in proportion to the amount of $^{137}\text{Cs}$ introduced.

By the measurement of size distribution of fine soil particles, it was revealed that the concentration of $^{137}\text{Cs}$ in the effluent decreased exponentially with the length of layer in the case of monodispersed fine soil particles.

From these results, the first-order irreversible reaction formula was obtained in order to formulate the reaction of $^{137}\text{Cs}$ adsorbed on fine soil particles with soil layer. A good agreement was obtained between the predicted result by polydispersed irreversible model and experimental one.

KEYWORDS: radionuclide migration, cesium 137, unsaturated soil layer, fine soil particle, particle size, filtration, irreversible model

I. INTRODUCTION

Some researches have been performed on the migration behavior of radionuclides in a soil layer to assess the long-term risk of the shallow land disposal of low level radioactive wastes\(^{(1)\sim(5)}\). These researches are concerned in the development of laboratory simulation techniques for studying radionuclide migration in natural environmental conditions and in the development of mathematical interpretative models for the description of involved mechanisms.

In the modeling of the migration of radionuclides in a soil layer, it is often assumed that the interaction of radionuclide, such as $^{60}\text{Co}$, $^{90}\text{Sr}$ and $^{137}\text{Cs}$, with soil can be described with equilibrium theory. And researchers reported that the migration of $^{85}\text{Sr}$ or $^{90}\text{Sr}$ could be predicted by using the distribution coefficient ($K_d$)\(^{(6)\sim(8)}\). Although the migration of a radionuclide which is mainly in an ionic form can be predicted by using the distribution coefficient, some researchers reported that small amounts of $^{60}\text{Co}$ and $^{137}\text{Cs}$ migrated much faster than the predicted migration using distribution coefficients\(^{(9)\sim(11)}\). Those are considered to be particulate forms, like cobalt hydroxides ($^{60}\text{Co}$ ($\text{OH}_2$) and $^{137}\text{Cs}$ adsorbed on fine soil particles.

Ohnuki et al.\(^{(12)}\) studied the migration of $^{137}\text{Cs}$ in soil layer, and found that the most of $^{137}\text{Cs}$ contained in the effluent was one adsorbed on fine soil particles and the migration could be dealt with several retardation factors corresponding to the diameter of fine soil particles on which $^{137}\text{Cs}$ was adsorbed. However, it is seemed that their model is not sufficient, because they assumed that equilibrium theory describes the interaction between fine soil particles and soil without the identification of reaction formula.

Saltelli et al.\(^{(13)}\) and Travis et al.\(^{(14)}\) have

* Tokai-mura, Ibaraki-ken 319-11.
studied the migration of colloidal ${}^{241}$Am (smaller than some micro-meter in diameter) in a soil layer, and found that the migration of ${}^{241}$Am predicted based on filtration theory agreed well with the experimental results. Since fine soil particles on which $^{137}$Cs is adsorbed consist mainly of clay (smaller than 2 $\mu$m in diameter), the migration of $^{137}$Cs adsorbed on fine soil particles in a soil layer seems to be described with filtration theory.

The aim of this study is to achieve a quantitative description of the migration of $^{137}$Cs adsorbed on fine soil particles in a soil layer. The results of migration experiments, the time dependent concentration of $^{137}$Cs adsorbed on fine soil particles in the effluent through a sandy soil layer and the concentration profile in the soil layer, are analyzed with filtration theory.

II. EXPERIMENTAL

1. Experimental Apparatus

The experimental apparatus as schematically shown in Fig. 1 is composed of soil column, micro-tube pump, fraction collector, etc. The column made of polyvinyl chloride was 5 cm in inner diameter and 25 cm in height. Soil packed in the column was coastal sand collected at the site of Tokai Research Establishment, Japan Atomic Energy Research Institute, which was sieved to remove gravel soils larger than 1,680 $\mu$m in diameter and had the density of 2.6 g/cm$^3$ (mean diameter of 350 $\mu$m). The soil column was prepared by putting glass beads (1,500 $\mu$m in diameter) to improve drainage, and then by pouring the soil to make a bed thickness of 5, 10 or 20 cm.

The suspension of fine soil particles on which $^{137}$Cs was adsorbed was prepared as follows. About 3 g of soil particles smaller than 37 $\mu$m in diameter, which was obtained by sieving coastal sand using JIS-standard sieve, was mixed with distilled water in the glass bottle of 10 cm in height. After sedimentation for 1 h (the particles larger than 5.6 $\mu$m in diameter were removed during sedimentation), the supernatant containing fine soil particles was taken out and radioactive aqueous solution containing $^{137}$Cs of about 33 $\mu$Ci as $^{137}$CsCl was charged into the supernatant. The suspension was diluted to the volume of 2,000 ml with distilled water. Then, fine soil particles on which $^{137}$Cs was adsorbed were separated from the suspension by centrifugation (3,000 rpm $\times$ 10 min $\times$ 3 times) to remove ionic $^{137}$Cs. The separated fine soil particles were suspended in distilled water of 2,000 ml and the concentration of $^{137}$Cs at the pouring portion into soil column was 1.2 $\times$ 10$^{-2}$ $\mu$Ci/ml. During the experiments, the suspension was stirred with a magnetic stirrer to prevent the sedimentation of fine soil particles.

2. Migration Experiments

Tritiated water was used to investigate the water flow in unsaturated soil layer. After making the steady water flow in the soil layer by pouring distilled water of about three times of pore volume, tritiated water of $1.1 \times 10^{-2}$ $\mu$Ci/ml was poured into the soil column at the constant feed rate of about 3 ml/min. The effluent was sampled with a fraction collector. Each 4 ml of the effluent samples was mixed with liquid scintillator (AQUASOL-2) of 12 ml and the concentration of $^3$H was determined by liquid scintillation counting technique.

The migration experiments were carried out as follows. After making the steady water flow in the soil layer by pouring distilled water of about three times of pore volume, the suspension of fine soil particles on which $^{137}$Cs was adsorbed was flowed into the soil column at the constant feed rate of about 3 ml/min. During the migration experiments, the effluent was sampled with...
a fraction collector. Then, each aliquot was adjusted to the volume of 20 ml to make the same efficiency of radioactivity measurement. After the experiment, the soil layer was cut into 1 cm sections to draw the concentration profile of 137Cs in the soil layer. Each 10 g of the dried soil samples was mixed with a hydrochloric acid of 45 ml (0.1 N) to maintain the homogeneity of radioactivity measurement. The concentrations of 137Cs in the effluent samples and in the soil samples were determined with a pure Ge semiconductor detector.

The experimental conditions are shown in Table 1.

<table>
<thead>
<tr>
<th>No.</th>
<th>Length (cm)</th>
<th>Porosity</th>
<th>Feed rate 3H (ml/min)</th>
<th>Feed rate 137Cs (ml/min)</th>
<th>Concentration 3H (μCi/ml)</th>
<th>Concentration 137Cs (μCi/ml)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>5</td>
<td>0.41</td>
<td>3.0(63')</td>
<td>3.0(197')</td>
<td>1.1×10^{-1}</td>
<td>1.2×10^{-2}</td>
</tr>
<tr>
<td>2</td>
<td>10</td>
<td>0.42</td>
<td>2.9(181')</td>
<td>3.0(210')</td>
<td></td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>20</td>
<td>0.41</td>
<td>2.9(181')</td>
<td>2.7(710')</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

† Total volume (ml)

### III. RESULTS AND DISCUSSION

The concentrations of 3H and 137Cs in the effluent are shown in Fig. 2 (a), (b), respectively. The concentrations of 137Cs are gradually approaching the saturated values, similar to the case of the effluent of 3H. Although the saturated concentration of 3H in the effluent is independent of the length of layer and is the same value as that of poured tritiated water, that of 137Cs is dependent of the length of layer. The saturated concentration of 137Cs is 2.1×10^{-3} μCi/ml for the layer of 5 cm in length, 1.5×10^{-3} μCi/ml for the layer of 10 cm in length, 1.0×10^{-3} μCi/ml for the layer of 20 cm in length, respectively. It is found that a soil layer reduces the concentration of 137Cs adsorbed on fine soil particles and the effect increases with the length of layer.

The concentrations of 3H and 137Cs in the effluent normalized by each saturated concentration described above are shown in Fig. 3. As can be seen from Fig. 3, the concentration distribution of 137Cs is similar to that of 3H, which is independent on the length of layer. It is obvious that a soil layer do not retard the migration of 137Cs adsorbed on fine soil particles and 137Cs adsorbed on fine soil particles migrates with the same velocity as water flow.

#### Fig. 2 (a) Concentration of 3H in effluent

#### Fig. 2 (b) Concentration of 137Cs in effluent
Fig. 3 Normalized concentrations of $^3$H and $^{137}$Cs in effluent

The concentration profiles of $^{137}$Cs in the soil layer are shown in Fig. 4. The concentration of $^{137}$Cs in the layer upper than 5 cm in depth increases in proportion to the increase in the amount of $^{137}$Cs introduced.

The reaction of a radionuclide in a soil layer is often described with equilibrium theory, and it is known that a soil layer retards the migration of the radionuclide, but does not reduce the saturated concentration of the radionuclide in the effluent. However, the experimental results mentioned above are different from those predicted by equilibrium theory. These suggest that the reaction of $^{137}$Cs adsorbed on fine soil particles in a soil layer might not be described with equilibrium theory.

The size of fine soil particles used appears to be distributed continuously and the reaction of fine soil particles with soil layer may depend upon the size of fine soil particles. For simplifying consideration, the concentration of $^{137}$Cs adsorbed on fine soil particles in the initial suspension and in the effluent were measured after sedimentation of fine soil particles (2, 8 and 32 h), and the size distribution of fine soil particles was determined by gravitational sedimentation method\(^{(13)}\).

The relation between the concentration of $^{137}$Cs and the length of layer is shown in Fig. 5 for four ranges of particle diameter. In Fig. 5, the diameter of 0.827, 1.65 and 3.31 $\mu$m is 100% cut-off-diameter corresponding to the sedimentation time of 32, 8 and 2 h, respectively. The particles whose ranges of diameter are 0.827 to 1.65 $\mu$m and 3.31 to 5.6 $\mu$m are removed by the layer of 5 cm in length. However, the particles whose ranges of diameter are $<$0.827 $\mu$m and 1.65 to 3.31 $\mu$m pass through all the layers. Particularly, even the layer of 20 cm in length remove only a little of the particles whose range of diameter is $<$0.827 $\mu$m. These are seemed to be caused by the difference of particle size, because large particles are easily trapped by the micro-pore through which smaller particles can pass. In the latter two cases, the decrease of $^{137}$Cs concentration with the length of layer is linear in the semi-logarithmic figure. In the case where particle size is uniform (monodispersed case), it
is found that the concentration of $^{137}$Cs adsorbed on fine soil particles decreases exponentially with the length of soil layer and the decrease rate changes with the diameter of particles on which $^{137}$Cs is adsorbed.

In general, one-dimensional solute migration in a soil layer with water movement is given by the following equation (1), and the solute concentration is obtained by substituting the reaction formula between the solute and the soil layer into

$$fs(\partial C/\partial t)+(1-f)\rho(\partial Q/\partial t)=fsD(\partial^2 C/\partial X^2)-fsV(\partial C/\partial X), \quad (1)$$

where $f$: Porosity  
$s$: Degree of saturation  
$\rho$: Density of soil  
$D$: Dispersion coefficient  
$V$: Velocity of water  
$C$: Solute concentration in water  
$Q$: Concentration of solute adsorbed on soil layer  
$t$: Time  
$X$: Length.

Since $^{137}$Cs adsorbed on fine soil particles is transported by water, the transport is expressed by Eq. (1).

In the part where the concentration of $^{137}$Cs in effluent is saturated, $\partial C/\partial t$ equals to zero, and $C$ is represented by $a \exp(-bX)$ ($a$ and $b$ are constant) since the concentration in effluent decreases exponentially with the length of layer (Fig. 5). Substituting those conditions into Eq. (1), the following equation is obtained:

$$(1-f)\rho(\partial Q/\partial t)=fs(D\beta^2+V\beta)C. \quad (2)$$

Thereby, the reaction of $^{137}$Cs adsorbed on fine soil particles with soil layer is expressed by the following first-order irreversible reaction formula ($K$ is constant):

$$\partial Q/\partial t=KC. \quad (3)$$

This equation is equal to filtration equation (13), and the reaction of $^{137}$Cs adsorbed on fine soil particles with soil layer might be described by filtration theory. Dividing the value of the concentration of $^{137}$Cs in soil layer (Fig. 4) by the experimental time, the mean value of $\partial Q/\partial t$ at the soil surface during each experiment is calculated to be $4.9 \times 10^{-4} \mu$Ci/g·min for the layer of 5 cm, $5.0 \times 10^{-4} \mu$Ci/g·min for the layer of 10 cm, $4.7 \times 10^{-4} \mu$Ci/g·min for the layer of 20 cm, respectively. These results are consistent with Eq. (3), because $C$ is a constant ($1.2 \times 10^{-2} \mu$Ci/ml) at the soil surface.

The parameters required to predict the migration of $^{137}$Cs adsorbed on fine soil particles in a soil layer, such as water flow velocity, dispersion coefficient, reaction constant ($K$), are determined from the experimental results, and the concentration of $^{137}$Cs in the effluent and in the soil layer are calculated from Eqs. (1) and (3).

Water flow velocity and dispersion coefficient of radionuclide are determined using non-reactive tracer, such as anion or $^3$H. In this study, they are determined from the time dependent concentration of $^3$H in the effluent (shown in Fig. 2(a)). It is known that the concentration of $^3$H in effluent is given by the following equation (16) ($C_0$ is the initial concentration):

$$C=0.5C_0[\exp(VX/D)erfc{(X+Vt)/2\sqrt{Dt}}+erfc(X-Vt)/2\sqrt{Dt}]. \quad (4)$$

The water flow velocity and the dispersion coefficient were determined by the least square data fitting of Eq. (4) to the measured concentration distribution of $^3$H in the effluent. The results are shown in Table 2 together with the degree of saturation calculated from porosity, feed rate of suspension, cross-sectional area of column and water flow velocity.

Table 2 Values of flow velocity, dispersion coefficient and degree of saturation

<table>
<thead>
<tr>
<th>No.</th>
<th>1</th>
<th>2</th>
<th>3</th>
</tr>
</thead>
<tbody>
<tr>
<td>Flow velocity (cm/min)</td>
<td>0.43</td>
<td>0.47</td>
<td>0.64</td>
</tr>
<tr>
<td>Dispersion coefficient (cm$^2$/min)</td>
<td>0.064</td>
<td>0.074</td>
<td>0.53</td>
</tr>
<tr>
<td>Degree of saturation</td>
<td>0.87</td>
<td>0.74</td>
<td>0.57</td>
</tr>
</tbody>
</table>

The reaction constant ($K$) is determined as follows: $\partial C/\partial t$ equals to zero in the part where the concentration of $^{137}$Cs in effluent is saturated. The following equation is obtained from Eqs. (1) and (3):

$$D(\partial^2 C/\partial X^2)-V(\partial C/\partial X)-\rho(KC/fs)=0. \quad (5)$$

The boundary conditions are $C=C_0$ ($X=0$) and...
and the initial condition is \( C = 0 \) for \( t \leq 0 \). The solution of Eq. (5) is written in

\[
C = C_0 \cdot \exp \left[ \sqrt{V + 4(1-f)\rho DK/s} X/2D \right].
\]

The reaction constants \( (K) \) were determined by the comparison of Eq. (6) with the concentration of \(^{137}\)Cs shown in Fig. 5. The results are shown in Table 3 (in this calculations, the fine soil particles of 0.827 ~ 1.65 \( \mu \)m and 3.31 ~ 5.6 \( \mu \)m in diameter assumed to decrease to the concentration of \( 10^{-4} \) \( \mu \)Ci/ml passing through the layer of 5 cm).

**Table 3** Reaction constant of \(^{137}\)Cs adsorbed on fine soil particles with soil layer

<table>
<thead>
<tr>
<th>Diameter (( \mu )m)</th>
<th>0~0.827</th>
<th>0.827~1.65</th>
<th>1.65~3.31</th>
<th>3.31~5.6</th>
</tr>
</thead>
<tbody>
<tr>
<td>( K ) (ml/g·min)</td>
<td>0.0012</td>
<td>0.014</td>
<td>0.018</td>
<td>0.085</td>
</tr>
</tbody>
</table>

The finite differential method (Crank-Nicolson method) was applied to each range of particle diameter for solving Eqs. (1) and (3). The total concentration of \(^{137}\)Cs adsorbed on fine particles in the effluent and in the soil layer were obtained by adding each concentration together. The results are shown in Fig. 2 (b) and Fig. 4 as solid line (the layer of 5 cm), broken line (the layer of 10 cm) and dotted line (the layer of 20 cm). Although the calculated concentration curve of \(^{137}\)Cs in the layer of 5 cm is slightly over-estimated as seen in Fig. 2 (b), the other concentration curves agreed fairly well with the measured curves. The migration of \(^{137}\)Cs adsorbed on fine soil particles is found to be predicted by using the first-order irreversible formula as a reaction formula.

### IV. CONCLUSIONS

To achieve a quantitative description of the migration of \(^{137}\)Cs adsorbed on fine soil particles in a soil layer, the suspension of fine soil particles on which \(^{137}\)Cs was adsorbed was poured into unsaturated sandy soil layers, and the concentration of \(^{137}\)Cs in the effluent and in the soil layer were determined. The following results were obtained:

1. A soil layer reduced the concentration of \(^{137}\)Cs in effluent, but did not retard its migration.
2. The concentration of \(^{137}\)Cs remained in the soil layer increased in proportion to the amount of \(^{137}\)Cs introduced.

From the measurement of the size distribution of fine soil particles on which \(^{137}\)Cs was adsorbed, it was found that the concentration of \(^{137}\)Cs in the effluent decreased exponentially with the length of layer in the monodispersed case. The first-order irreversible reaction formula was obtained in order to formulate the reaction of \(^{137}\)Cs adsorbed on fine soil particles with soil layer. The concentration of \(^{137}\)Cs in the effluent and in the soil layer calculated from the reaction formula for each size of fine soil particles agreed well with experimental values. It was confirmed that the migration of \(^{137}\)Cs adsorbed on fine soil particles in a soil layer could be predicted by using the first-order irreversible reaction formula.

### ACKNOWLEDGMENT

The author would like to thank Drs. Y. Wadachi, T. Yamamoto and Mr. H. Matsuzuru of Japan Atomic Energy Research Institute for the helpful discussion.

### REFERENCES