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SAFETY STUDY ON GROUND DISPOSAL OF LOW-LEVEL WASTES

1983

DIVISION OF ENVIRONMENTAL SAFETY RESEARCH

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#### Introduction

The ground disposal of low-level wastes is considered as one of the most potential alternative methods. The ground disposal has been thought to be inadequate because of high density of population and limited terrain in Japan. In the case of sea dumping, however, there are several limitations concerning the amount and kind of wastes to be able to dump, which are specified by so-called the London Convention. The role of ground disposal in the management of low-level wastes may inevitably be essential in present.

To realize the ground disposal, it is necessary to grasp the migration of radionuclides throughout a geosphere, and to estimate the radiation dose to which the public would be exposed. This research program aims at evaluation the migration behavior of radionuclides in the aerated and aquifer zones under the condition simulating disposal site, so that basic informations may be obtained to achieve the comprehensive safety assessment of the disposal.

The migration of <sup>137</sup>Cs, <sup>60</sup>Co, <sup>90</sup>Sr, <sup>85</sup>Sr has been studied by using several apparatuses which can model aerated and aquifer zones. Furthermore, to obtain more realistic data concerning the migration and distribution of radionuclides, the facility of "Simulation Test for Environmental Radionuclide Migration" is now under construction.

## (1) Past progress

 (a) Adsorption behavior and distribution coefficients of radionuclides<sup>1</sup>)~3)

The adsorption behavior of radionuclides on sand and the influence of pH and concentration of radionuclides on the distribution coefficient( $K_d$ ) have been studied in advance. Sand used here (coastal sand collected from the site of JAERI) was subjected to washing and screening to get the sand sample of mode radius 350  $\mu$ m, density 2.63 g/cm<sup>3</sup>. All of equilibrium measurements between sand(10 g) and radioactive aqueous solution(200 ml) were made by a batch method.

As seen in Fig.1,  $^{137}$ Cs has relatively high K<sub>d</sub> values over wide range of pH and gives the maximum K<sub>d</sub> in neutral region, while the value of K<sub>d</sub> for  $^{85}$ Sr is lower than that for  $^{137}$ Cs and has the maximum K<sub>d</sub> in alkaline region. The increasing in K<sub>d</sub> with the increasing pH may be explained by decreasing the competitive action of H<sup>+</sup> ion in the adsorption process, provided that this process in a low pH range is mainly attributed to ion exchange.

It is also found that adsorption of these cationic radionuclides on sand is approximately described by the Henry equation,

### $X = K_d C$

where X is the equilibrium amount adsorbed (mol) and C is the equilibrium concentration (mol/l).

(b) Migration and distribution of radionuclides in aerated and aquifer zones<sup>4</sup>)~7)

In series of fundamental studies on safety evaluation of the ground disposal of low-level wastes, the migration and distribution of <sup>137</sup>Cs, <sup>60</sup>Co, <sup>90</sup>Sr, <sup>85</sup>Sr in aerated and equifer

zones (sand) were examined by using medium-scale soil zone model apparatuses (Fig. 2 and Fig. 3). The migration and distribution of radionuclides were obtained by measuring the radioactivity of effluent passed through the outlet of soil column or soil vessel and sand collected from the column or the vessel.

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Figure 4(a), (b), (c) show the two dimensional distribution of radionuclides in aerated sand layer. The distribution of  $137_{\rm CS}$  is relatively uniform in both vertical and horizontal directions of the sand. That of  $^{60}$ Co is particularly large in vertical direction. And the distribution pattern of  $^{85}$ Sr lies midway between those of  $^{137}$ Cs and  $^{60}$ Co. It is found from these distribution curves given in the figure that the migration velocity follows the order;

## 137 cs < 60 co < 85 sr

These migration velocity order may be explained by the chemical behavior of each nuclide. Namely,  $^{137}$ Cs exists as cation in the solution, thus it can be adsorbed on sand by the ion exchange reaction, that results in the lowest migration velocity. On the other hand, a part of  $^{60}$ Co hydrolyzes to form an insoluble hydroxide,  $^{60}$ Co(OH)<sub>2</sub> which cannot be contributed to the ion exchange adsorption, therefore this hydroxide migrates downward. Since  $^{85}$ Sr also exists as cation in the solution, it can contribute to ion exchange adsorption reaction. This process, however, is largely affected by co-existent calcium ion and carbonate ion. That probably results in the highest migration velocity.

Migration and distribution of  $^{137}$ Cs,  $^{60}$ Co,  $^{85}$ Sr in aquifer sand layer also have been studied. As a result,  $^{137}$ Cs gives unexpectedly large migration in the sand. To elucidate this phenomenon, a few additional experiments have been carried out by means of small-scale soil column. Consequently, it is found that fine silt adsorbs <sup>137</sup>Cs tightly and it migrates in the sand layer according to water flow.

(2) Recent progress

(a) Prediction of radionuclide migration in aerated zone<sup>7)</sup>, 8) To predict the radionuclide migration in the aerated sand layer,

the following solution of one-dimensional transport-dispersion equation proposed by Y. Inoue, et al.<sup>9)</sup> is used for the calculation;

$$\frac{q}{q_0} = \frac{1}{2} \left\{ 1 - \operatorname{erf}\left(\frac{x - vt/Kf}{2 \int D_m vt/Kf}\right) \right\}$$

where, x : depth of aerated zone (cm)

v : velocity of water movement (cm/min)

D<sub>m</sub>: standard diffusion constant (cm)

t : feeding time of radioactive solution (min)

K<sub>f</sub>: retardation factor

In comparison with the calculated and experimental data, both satisfactorily coincides each other in case of migration of <sup>85</sup>Sr. However, the migration <sup>60</sup>Co and <sup>137</sup>Cs can only be predicted in a conservative way. We are now going to investigate the predictive equation for the migration of <sup>137</sup>Cs, particularly, in consideration of silt migration.

(b) Measurement methods for velocity of water movement in aerated and aquifer zones 10), 11)

In order to develop the useful measurement method for the velocity of water movement, fast neutron radiation method for

aerated zone and pulse geating method for aquifer zone have been studied, respectively.

Fast neutron radiation method is based on detecting the strength change of fast neutron when passes through soil layer containing water. This measurement system consists of <sup>252</sup>Cf as neutron source and NE-213 scintillator as detector. Figure 5 shows the working curves of fast neutron radiation method about the aerated sand layer. All of curves give the linear relationship between decreasing ratio of counting (1/min) and velocity of water movement (cm/min). In result, it is found that the sensitivity of measurement increases as the diameter of soil column increases.

Pulse heating method is to detect the change of thermal conductivity, that is, temperature change of water when moves through soil layer. This measurement system has pulse heating mechanism and thermistor type sensor. Figure 6 shows the working curves of pulse heating method about the aquifer sand layer. All of curves give the good linear relationship between tempera ure increase of water (°C) and velocity of water movement (mm/min). It is therefore found that the most adequate space between heater and sensor is 10 mm.

# (3) Future plan<sup>11)</sup>, 12)

Japan Atomic Energy Commission has published the disposal principles of radioactive wastes, that is, low-level wastes should be disposed of by sea dumping and burial in ground. In order to contribute the safety assessment of ground disposal of low-level wastes arising mainly from nuclear power station, an extensive test on radionuclide migration is now being planned by using large-scale apparatus. That apparatus can simulate a shallow land of disposal site, thus this migration test is the useful alternative of field test using radionuclides.

Based on the information obtained with such migration test, we are able to evaluate the retardation abilities of aerated and aquifer zones as natural barrier, and moreover, to establish the migration model for safety assessment of disposal site.

This research program is called "Simulation Test for Environmental Radionuclide Migration (STEM)". The facility is now under construction (see Fig. 7) and according to the time schedule, the radionuclide migration test will be strated in the end of FY 1983 after the preliminary test (see Table 1).

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Fig. 2 Aerated zone model apparatus. Soil column : 30 cm \$ x 120 cmh.



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Fig. 4 Two dimentional distribution of radionuclides in aerated zone (sand).



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## Table 1 Time schedule of simulation test for environmental radionuclide migration.

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