

## EFFECT OF Ca IONS IN SOLUTION TO Cs-137 SORPTION INTO SOIL SAMPLES

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### ABSTRACT

EFFECT OF Ca IONS IN SOLUTION TO Cs-137 SORPTION INTO SOIL SAMPLES. Experiment of effect of Ca ions exist in solution to sorption of Cs-137 radionuclide into soil samples have been done. In environment, Ca ion is a major constituents of mineral exists in a solution, also this mineral can come from the weathering of waste matrix/packages, and soil as a host of the radwaste disposal facility has an important role to retard radionuclide migration such as Cs-137 from the facility into biosphere by sorption mechanism. The objective of work is to study the effect of Ca ion co-existing in a solution to Cs-137 sorption into soil samples. The experiments were evaluated by determine the sorption kinetic, sorption isotherm and effect of Ca ions to sorption of Cs-137 into soil. Batch method was performed for the laboratory work and Cs-137 radionuclide was used as a tracer. Concentrations of CsCl is  $10^{-4}$  M and  $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$  as a  $\text{Ca}^{2+}$  simulated ions were ranged from  $10^{-6}$  to  $10^{-2}$  M. The results showed that saturation condition of Cs-137 into soil sample was reached after 2 days contact time, with  $K_d$  was 4500 ml/g. Sorption isotherm of Cs-137 into soil followed *Freundlich law*, and the existence of Ca ions in a solution was not give an effect to the sorption of Cs-137 into soil samples.

Keywords: *Ca ions, Cs-137, Sorption*

### INTRODUCTION

For the issue of disposal method for low-level radwastes (LLW), International Atomic Energy Agency (IAEA) have recommend to its member states to dispose this kind of waste into a near surface disposal facility [1]. Immediately after closure period, the scenario of radionuclide (RN) migration into biosphere through groundwater movement becomes occur. Groundwater around the facility will penetrate into facility through several types of engineering barrier such as concrete wall, buffer material, waste container, waste matrix. For long-term period of storage, groundwater will reach and contact to RNs contained waste packages and some of RNs will dilute to groundwater. The soluble RN from waste packages then follow together with groundwater movement and release to environment or biosphere. Soil as a host of disposal facility and as a natural barrier has an important role to retard the migration of RN into environment through sorption mechanism. From the safety point of view, the soil is an important part of radwaste disposal system. Based on the NEWJEC, and internal study, Lemahabang area in Muria Peninsula-Central Jawa is a most potentially site candidate for first Indonesian nuclear power plan and one of potential site candidate for radwaste disposal facility [2,3] and soil samples for the experiment were taken out from these area.

Radiocesium (Cs-137) is one of most considered RNs on LLW disposal system due to its half-life (30 y) and also as a dominant nuclide in an inventory of LLW [4]. The existence of metal ions in groundwater is another problem to be considered. Calcium ions were a major soluble constituent of mineral in groundwater ( $10^{-6}$  to  $10^{-3}$  M) [5] and it also can come from the degradation of solidification/immobilization (cement) results of waste matrix/packages or weathering of bentonite minerals as buffer materials in the disposal facility [6]. In the radionuclide-soil interaction, it is important to estimate the effect of competing cations/metal ions originally present in the groundwater (such as Ca ions). Due to the previous condition, Ca ions as co-existence and competing ions in groundwater are predicted to give an effect to the sorption of Cs-137 into soil samples and the study of Ca ions co-existence effect to Cs-137 sorption into soil samples become an important study.

The objective of the experiment is to study the effect of co-existence ions (Ca ions) in a solution to Cs-137 sorption into soil samples. The experiment was performed by evaluate the kinetic, sorption isotherm and effect of co-existence of Ca ions in solution. The experiment has a link with the experiments of environment safety around the disposal facility, and the obtained results could be contributed to environment safety assessment stage for emergency planning of disposal facility in the future.

**EXPERIMENTS**

**Materials**

Soil samples were taken out from Lemahabang area, Muria Peninsula-Central Jawa. After cleaned-up and separated from roots, gravels and leaves, soils were crushed and sieved into 100 mesh particle size. Dried with oven (T 100 °C, 4 h), and then samples were stored in a desiccator for a week. The experiments were performed by contacting the soil with CsCl labeled with Cs-137,  $C_{CsCl}: 10^{-8}$  M, sp. activity of Cs-137 is  $10^{-2}$   $\mu$ Ci/ml by using rolling method. Batch method was performed for the laboratory work and Cs-137 radionuclide was used as a tracer. Concentration of Ca were made by diluting  $CaCl_2 \cdot 2H_2O$  in demineralized water and ranged from  $10^{-6}$  to  $10^{-2}$  M.

**Procedures**

Kinetic,

Soil samples were contacted with demineralized water in a 20 ml volume of PE vial and equilibrated for 3 days. Add Cs-137, CsCl ( $10^{-8}$  M) and shaking them up to 7 days, ratio of solid-liquid was  $10^{-2}$  g/ml as conducted in ref.[7]. Periodically the solution was taken out from the vial for measured its  $\gamma$ -activity. The mixtures were separated by using centrifuge apparatus *Marathon 21K Fisher Scientific* (2500 rpm, 10 min.), then 1 ml of aliquot was taken out and measured its  $\gamma$ -activity by using a liquid scintillation counter (LSC) *Tri-Carb Packard Canberra 1600-TR* and Cs-137 distribution ( $K_d$ ) was calculated by using equation [8],

$$K_d = \frac{C_s}{C_l} = \frac{A_0}{A} - \frac{V}{W} \text{ ml / g} \tag{1}$$

where Cs and Cl are RN's concentrations in solid (mol/g) and liquid (mol/ml),  $A_0$  and A are initial and final activities of RN, V is volume (ml) and W is solid mass (g).

Sorption Isotherm,

The solution contains CsCl  $10^{-4}$  M and Cs-137 were contacted with soil samples and then shaking the mixture by rolling method for 3 days. Separation and calculation methods were similar with previous method.

Ca ions co-existence,

The experiment is to simulate the effect of Ca ions co-existence in groundwater to Cs-137 sorption into soil samples. Concentration of  $CaCl_2 \cdot 2H_2O$  were ranged from  $10^{-6}$  to  $10^{-2}$  M,  $C_{CsCl}$  was  $10^{-4}$  M. Soil, Ca ion and CsCl/Cs-137 were mixed in a 20 ml volume of PE vial and then rolled for 4 days. Separation and calculation methods were similar with previous experiments.

**RESULTS AND DISCUSSIONS**

The result of sorption kinetic of Cs-137 onto soil is shown in Fig. 1. The distribution coefficients of Cs-137 reached equilibrium phase after 2 days contacting with  $K_d$  value was 4500 ml/g. Equilibrium conditions were indicated with  $K_d$  values become constant after 2 days contact time, even contact time was continued up to 7 days ( were indicated with a plateau line). In the equilibrium condition, RN sorbed onto soil also was considered has saturated and equilibrium from solution into soil *vice versa*.

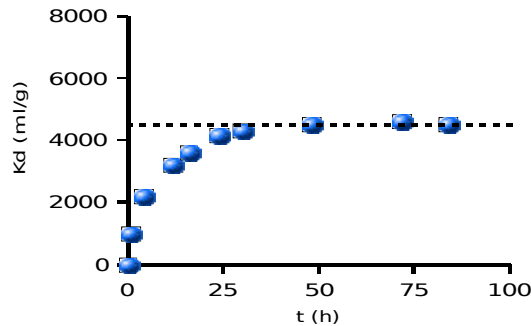


Fig. 1 Kinetics of Cs-137 sorption onto soil

The result of isotherm sorption experiment was shown in Fig. 2. Plateau in the curve was adopted as a sorption capacity value of soil,  $6.92 \times 10^{-4}$  meq/g with pH of solution was around 7. This situation was indicated with the concentration of Cs-137 in soil ( $C_s$ ) still constant even Cs-137 in solution ( $C_i$ ) increased. Straight line of slope is indicating that the isotherm sorption process followed *Freundlich law* with equation [9],

$$C_s = C_a(K.C_i)^n \tag{2}$$

and in a logarithmic style the equation (2) becomes,

$$\text{Log } C_s = \text{Log } C_a + n \text{ Log } K + n \text{ Log } C_i \tag{3}$$

where  $C_a$  and  $K$  are sorption capacity and equilibrium constant, and slope ( $n$ )= 0.509. By inserting  $C_s$ ,  $C_i$ ,  $C_a$  and  $n$  into eq.(3), from calculation we obtain the equilibrium constant ( $K$ ) becomes  $6.87 \times 10^{-4}$  meq/ml.

The result from the experiment of effect of Ca ions to Cs-137 sorption onto soil samples was shown in Fig. 3. Addition of Ca ion into solution contained RN does not give any significant effect to Cs-137 sorption onto soil, due to cesium ions does not change its speciation in solution [10], and the result of the experiments give an identically result for both results (with and without Ca ions co-existence).

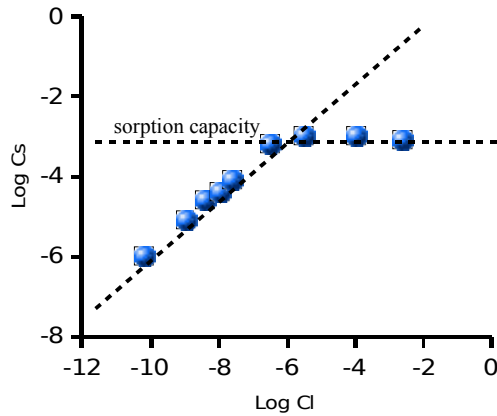


Fig. 2 Isotherm of Cs-137 sorption onto soil

Effect of  $\text{Ca}^{2+}$  concentrations to sorption of Cs-137 was shown in Fig.4. The changing of Ca concentrations does not give any influence to Cs-137 sorption due to differentiation of their ion size and mobility of ions.

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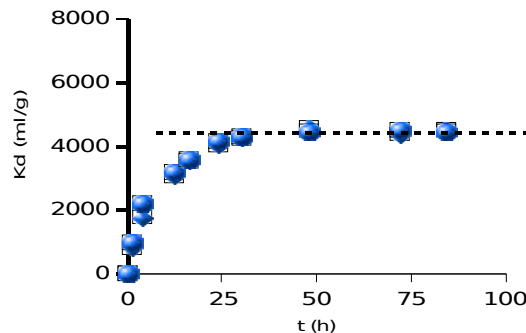


Fig. 3 Effect of Ca ions to Cs-137 sorption

**CONCLUSION**

Experiment of effect of Ca ions exist in solution to sorption of Cs-137 radionuclide into soil samples have been done. The results showed that equilibrium sorption was reached after 2 days contacted with distribution coefficient,  $K_d = 4500 \text{ ml/g}$  with pH 7. Sorption capacity of soil was  $6.92 \times 10^{-4} \text{ meq/g}$  and  $n = 0.509$ . Addition of Ca ion into solution contains RN does not give any significant effect to Cs-137 sorption onto soil, indicated by obtaining data give an identically data for both results (with and without Ca ions co-existence).

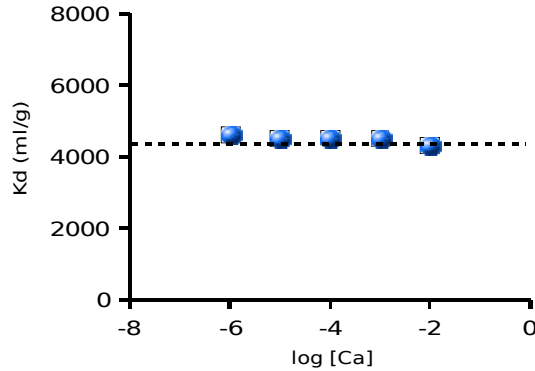


Fig. 4 Effect of  $C_{ca}$  to Cs-137 sorption

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