

Study of Distribution Coefficient of ^{90}Sr on Citumbuk Bentonite and Sukaramai Kaolinite

Studi Koefisien Distribusi ^{90}Sr pada Bentonit Citumbuk dan Kaolin Sukaramai

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ABSTRACT

The study of distribution coefficient of strontium on bentonite and kaolinite samples, collected from Citumbuk, Bogor and Sukaramai, Padang, has been investigated under batch experiments. The sorption ratio was determined as a function of time, ionic strength and Sr concentration in the solution. The strontium was extensively absorbed in the samples, with Kd values of 13,700 and 1,600 mL/g for bentonite and kaolinite samples respectively. The other results showed that ionic strength of the solution and initial concentration of Sr have given an effect to ^{90}Sr sorption onto samples.

Keywords: buffer materials, ^{90}Sr , Kd

ABSTRAK

Studi tentang koefisien distribusi dari stronsium pada sampel bentonit dan kaolin yang diambil dari lokasi Citumbuk, Bogor dan Sukaramai, Padang, telah diteliti secara catu. Rasio penyerapan ditentukan sebagai fungsi waktu, kekuatan ionik dan konsentrasi Sr di larutan. Stronsium telah terserap dengan baik pada sampel, dengan nilai Kd adalah 13.700 dan 1.600 mL/g masing-masing untuk sampel bentonite dan kaolin. Hasil lainnya menunjukkan bahwa kekuatan ionik dan konsentrasi awal Sr di larutan telah memberikan pengaruh pada penyerapan ^{90}Sr ke dalam sampel.

Kata kunci: bahan penyangga, ^{90}Sr , Kd

INTRODUCTION

One of the predominant long-lived radionuclides as a high yield product of nuclear fission is ^{90}Sr and it is become an important component of radioactive waste with 28 years half-life and emitting pure β -ray. When ^{90}Sr releases into the environment and inserted into human body through the food chain, Sr can substitute Ca^{+2} ion in human bone structure and increase the risk of diseases such as leukemia [1].

Like cassium, strontium has a very high uptake capacity when they interact with the material, so their sorption in to the soil can retard their migration. In the present work, we have studied the sorption behavior of ^{90}Sr into bentonite and kaolinite materials as a function of time, ionic strength, and SrCl_2 concentration.

Previous studies have been carried out on geological materials such as clay and soil to ^{90}Sr [2][3][4]. The results showed

that the sorption ratio of ^{90}Sr is high enough, especially at low concentrations. However, for ^{90}Sr sorption onto quartz, apatite, calcite or iron oxides is very low ($K_d < 5$). These mainly due to the low ion exchange capacity of the minerals [5][6].

Bentonite and kaolinite materials have generally been chosen as candidates for the buffer material on the radioactive waste disposal [7][8]. The function of buffer materials in a disposal facility is to control the water or surface water penetration into the disposal facilities, and also absorb radionuclide if any, which is carried out by water flow from facility to environment. While transported in buffer material, ^{90}Sr may be absorbed onto buffer material surfaces.

In any sorption study, it is very common to use coefficient distribution or K_d value as the sorption parameter or indicator of an occurrence of ^{90}Sr sorption into samples. The K_d value is valid only for a special set of conditions such as pH, concentration of competing ion and temperature. In this study, the condition is set with no variation of pH and no treatment of the soil. Objective of the experiment is to get the specific data of ^{90}Sr sorption into Indonesian local mineral samples.

METHODOLOGY

The reagent used in the experiments were $\text{SrCl}_2 \cdot 6\text{H}_2\text{O}$, NaCl from *E. Merck* and ^{90}Sr as a tracer with sp. activity 3.7 MBq/5 mL from *Eckert & Ziegler Isotope Production* diluted in 0.1 M HCl. Samples material were taken from Citumbuk, Bogor, West Java and Sukaramai, Padang, West Sumatera for bentonite and kaolinite samples respectively. The experiments use 100 mesh particle sized samples.

The static batch method was chosen for performing the experiment. The samples were

weighted using analytic scale unit, Mettler serie AE200, and mixed with the solutions contain $\text{SrCl}_2 \cdot 6\text{H}_2\text{O}$ in aquadest. Two series of samples (bentonite and kaolinite) for ^{90}Sr sorption experiment was prepared in a 20 mL PE vial. In the experiments of the function of time and ionic strength, the initial concentration used of $\text{SrCl}_2 \cdot 6\text{H}_2\text{O}$ was 10^{-8} M and for the experiment of SrCl_2 concentration loading, the initial concentration of ^{90}Sr was varied between 10^{-3} to 10^{-8} M $\text{SrCl}_2 \cdot 6\text{H}_2\text{O}$. Then each sample from each series labeled with ^{90}Sr . The ratio of solid: liquid phase was 10^{-2} g/mL.

Samples solutions were kept in polyethylene vials and then separation of the solid and liquid phases was carried out using centrifuge machine, Heraeus Labofuge 400 (10 min., 2500 rpm). The obtained aliquot of ^{90}Sr then evaluated with the detector system. Since strontium is a pure beta-emitter, the concentration of ^{90}Sr was measured by a liquid scintillation counter (LSC) unit, Perkin Elmer Tri-Carb 2910 TR.

One of the vials that contains ^{90}Sr without soil represents as a blank sample. This vial used to correct for any sorption on the walls of the vials. All samples were gently shaken in a rolling shaker at a low and constant speed (100 rpm). At several times, an aliquot from each vial series removed, separated and measured using LSC.

For desorption experiments, the solution was replaced by the fresh aquadest and then shaken again using a roller machine. The next procedure was similar to the previous experiment.

The sorption was described by distribution coefficient (K_d) of ^{90}Sr . K_d is the ratio of the number of ^{90}Sr sorbed in the solid phase to the number of ^{90}Sr remains in the solution. The concentration of strontium in the liquid and in the solid phase after

sorption was calculated from the measured activities as follows:

$$[C]_{eq} = \frac{A_f}{A_0} \cdot [C]_0 \quad (1)$$

$$[C]_{ads} = ([C]_0 - [C]_{eq}) \cdot \frac{V}{M} \quad (2)$$

where A_f and A_0 are the final and initial activity of ^{90}Sr in solution, $[C]_{eq}$ and $[C]_{ads}$ are the concentration of Sr in the liquid and the solid phase, respectively, $[C]_0$ is the initial concentration of Sr, V is the volume of solution (mL) and M is the dry mass of the sample (g).

RESULT AND DISCUSSION

The study of ^{90}Sr sorption as a function of time was carried out in a period over of 15 and 12 days for bentonite and kaolinite samples, respectively. Figure 1 shows the results of sorption as a function of contact time. The saturated condition reached after 12 and 5 days contact time with K_d values were 13,700 and 1,600 mL/g for bentonite and kaolinite samples respectively. The ^{90}Sr in the solution has reacted with the reaction site of the sample, and the number of Sr that is in solution become reduced bound to a solid phase. It goes on until the condition on the reaction site become saturated, indicated by the increase of K_d values. When the reaction site of the sample saturated with ^{90}Sr , the shape of the graph of K_d value becomes flat [9]. In the desorption experiments, displacement solution in vials with a fresh solution has led a portion of ^{90}Sr released from the samples and back to the solution. These treatment has increased the concentrations of Sr in the solution and the K_d values become decreased as showed in Figure 2. The equilibrium states were reached after 12 and 5 days shaking, and K_d values

are 5.800 and 630 mL/g for bentonite and kaolinite samples, respectively. The results indicated that a portion of ^{90}Sr bound strongly in samples and other ^{90}Sr released to solution. The reaction of ^{90}Sr and samples shows a reversible reaction [10].

NaCl is ionic metal constituents in groundwater media with quite high concentrations. The existence of NaCl concentration will interfere the sorption of ^{90}Sr into both samples. Figure 3 gives a clear view that the rising of NaCl concentration in solution can lower the K_d values of ^{90}Sr . Competition between Na and ions in solution were able to disturb the amount of ^{90}Sr sorbed into samples [11]. Sorption of ^{90}Sr onto samples was controlled with the competition of metal ions such as Sr^{2+} and Na^+ ions in solution.

The effects of loading concentration of SrCl_2 on the ^{90}Sr sorption onto bentonite and kaolinite samples are presented in Figure 4. It is observed that in the low concentration of SrCl_2 contributes higher K_d values for both samples, *vice versa*. The highest K_d value was found at the lowest Sr concentration due to the availability of reaction sites of the samples that will be saturated at higher concentration of SrCl_2 . In case of bentonite sample was showing superior sorption of ^{90}Sr compared with kaolinite due to the structure of the mineral [12]. Similar results have also been reported by Yildiz et.al and Galambos, et.al, however, they studied through ^{137}Cs isotope [12][13].

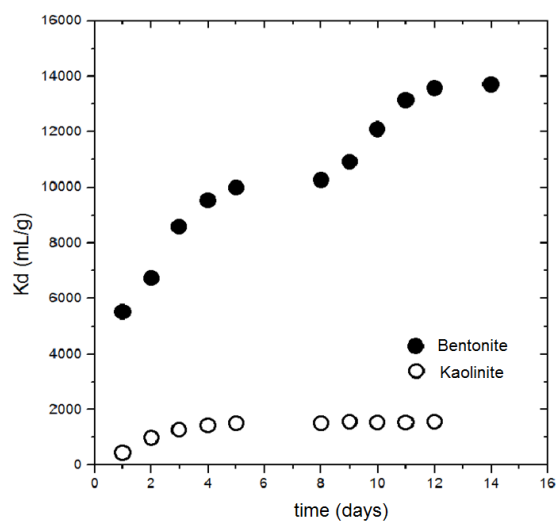


Figure 1. Sorption of ^{90}Sr in samples as a function of time.

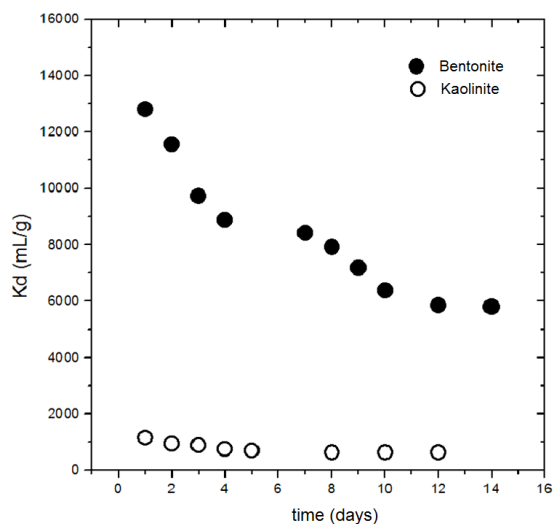


Figure 2. Desorption of ^{90}Sr from samples as a function of time.

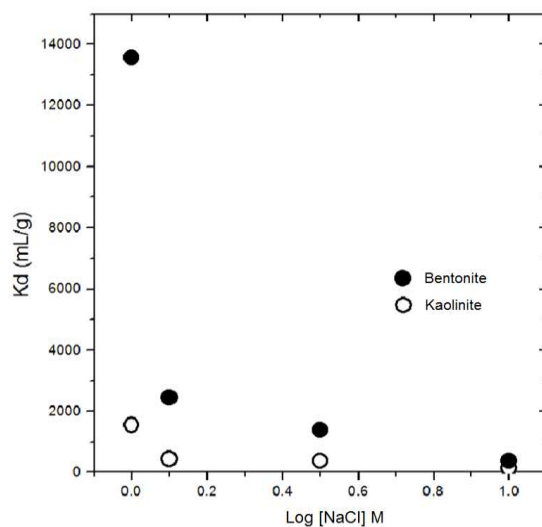


Figure 3. Sorption of ^{90}Sr as a function of NaCl concentrations.

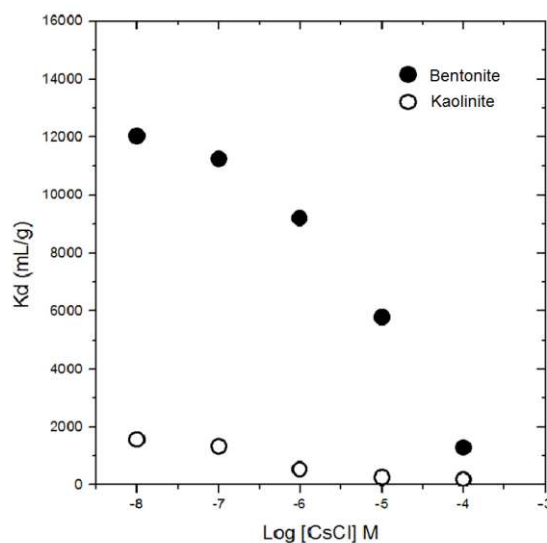


Figure 4. Sorption of ^{90}Sr as a function of SrCl_2 concentration

CONCLUSIONS

In the present work ^{90}Sr sorption into bentonite and kaolinite samples were investigated in a batch method. There are no variation of the pH and also no treatment of the samples. The sorption of ^{90}Sr into samples was very fast, assumed it was affected by ^{90}Sr concentrations in solution. A lower concentration of NaCl given a higher sorption of ^{90}Sr due to ions competition and a lower concentration of ^{90}Sr given a maximum sorption of ^{90}Sr in the samples due to more reaction sites on the samples were available.

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