Determination of diffusion coefficient of $^{137}$Cs at unsaturated zone of DH-2 site soil under $\delta = 1.41$ g.cm$^{-3}$ condition

Budi Setiawan and Nurul Efri Ekaningrum
Center for Radioactive Waste Technology-BATAN
Bld 71 3rd Floor, PUSPIPETEK, Serpong-Tangerang, Indonesia 15310

E-mail: bravo@batan.go.id

Abstract. Radiocesium is one of the reference radionuclide on low-level radioactive waste disposal system. For the safety assessment of environment around the disposal facility, it needs to be studied their characters on the candidate site of radioactive waste disposal facility. Diffusion coefficient is important to be studied especially on the unsaturated zone lies between the foundation of facility and the saturated zone. In this experiment, soil samples were taken out from the logging result of DH-2 hole of SP-4 site on the 7 m depth from surface. Work was done in the diffusion column by gravitation method under density condition was 1.41 g.cm$^{-3}$. The diffusion coefficient is determined in one dimension using non-steady diffusion method with variation of diffusion time and has obtained the smallest of Da value of $^{137}$Cs in the soil sample was $1.76 \times 10^{-12}$ m$^2$s$^{-1}$ at the 6 months of diffusion time. The results show that the longer of length diffusion time, the smaller diffusion value was obtained.

Keywords: $^{137}$Cs, DH-2 site, unsaturated zone, diffusion coefficient

1. Introduction

Radiocesium such as $^{137}$Cs together with $^{90}$Sr and $^{60}$Co are one of the reference radionuclides on the radioactive waste management system [1], due to their dominated on the radioactive waste inventory which has half-life $\leq 30$ years [2]. Radiocesium has half-life 30 years, easy associated with materials in groundwater and terrestrial, and easy to drift with groundwater and absorbed into food chain [3]. For that reason radiocesium should be studied their character after to be disposed in a disposal facility.

DH-2 site is one of the existing drilling points at the site candidate for the planned demonstration disposal facility construction in SP-4 location of Serpong Nuclear Center (SNC), Serpong-Tangerang, Banten, see Figure 1 [4]. The planning of disposal facility construction intended to complete the radioactive waste management facility at SNC. In SNC some existed radioactive waste management facilities such as chemical treatment, evaporation, incineration, compaction and interim storage facilities except disposal facility. Before we construct the disposal facility, safety assessment study should be performed, and one of important data should be found is the preparation of diffusion data of the site. Previously the sorption performance data of SP-4 location to radiostrontium has been studied [5], and also the determination of dry density of site soil has been done [6] where the dry density of soil was 1.41 g.cm$^{-3}$. The dry density value and then was used for the diffusion experiments. The diffusion phenomena usually occured on the unsaturated zone, in the SP-4 location was found at the 7 m depth from the soil suface, lies between the lowest part of foundation of facility and the saturated zone, see Figure 2 for the near surface disposal facility type [7].
Diffusion is the occurrence of material transfer through a media due to atomic or a particle movement due to presence of thermal gravitation. The diffusion processes can be irreversible. In the event of diffusion, concentration of material will change to time, \( \frac{C_x}{dt} \), and the material flux also the function of time. Figure 3 showed the changing of \( C_x \) concentration on the transient condition.

**Figure 1.** Location of planned demonstration disposal facility at Serpong Nuclear Center.

**Figure 2.** An illustration of disposal facility near surface type [7].
Figure 3. Diffusion at the transient condition.

The changing of concentration becomes the different between inlet flux at xa point and outlet flux at x point. The difference at time t is the changing of Cx concentration, and outlet flux will be approached by using Ficks II law,

$$\frac{dc_x}{dt} = D \frac{d^2 c_x}{dx^2}$$  \hspace{1cm} (1)

The total rate of concentration changing proportional to the Laplacian of initial concentration. For planar source with small amount of soluble substances in a cylinder system with infinite length, the equation (1) becomes,

$$C(x, t) = \frac{M}{2\sqrt{\pi Dt}} \exp \left( -\frac{x^2}{4Dt} \right)$$ \hspace{1cm} (2)

Where C is the total concentration of radiocesium (Bq), t is diffusion time (s), x is the distance of source with measurement point (m), and M is concentration of radiocesium per-square unit (Bq.m^2) [8]. And the equation (2) was changed in logarithmic form, the equation becomes,

$$\ln C = -\frac{x^2}{4Dt} + \ln \left[ \frac{M}{\sqrt{\pi Dt}} \right]$$ \hspace{1cm} (3)

And then the equation will be determined linearly followed the equation, \hspace{1cm} y = ax + b

Where y = ln C, \hspace{1cm} a = \frac{1}{4Dt} \hspace{1cm} and \hspace{1cm} x = x^2

Diffusion coefficient is obtained from direction coefficient of concentration C(x,t) as a function of distance x. The objective of the experiment is to determine diffusion coefficient of $^{137}$Cs on soil sample specimen of site candidate of demonstration disposal facility at SNC with dry density soil condition was 1.41 g.cm^-3.

2. Methodology
Soil samples used in the experiment was taken out from a logging result soil of drilled hole DH-2 at SP-4 location of SNC on the depth of 7 m from the surface. By assuming that the lowest part of
planned facility foundation at the depth of 6 m and aquifer zone is in the depth of 10 m from the surface, the location on the depth of 7 m placed at the unsaturated zone. And the diffusion coefficient is determined on the unsaturated zone.

Radiocesium, $^{137}$Cs solutions was ordered from Eckert & Ziegler Isotope Production with Sp. Activity 3.7 MBq/5 ml and the solution was dissolved to 10,000 Bq/ml as radioactive solution stock before used as a tracer in the experiments.

The one dimension of non-steady state diffusion experiment was performed in a diffusion cell as shown at Figure 4, in the dry density condition was 1.41 g.cm$^{-3}$ as a previous result [6].

![Figure 4](image.png)

Figure 4. Diffusion experiment in a diffusion cell, schematically [9].

Conditioning of soil sample was done by sinking method of soil sample in a diffusion cell in the demineralized water for 1 month. The conditioning was intended to saturate the soil sample specimen before the diffusion experiment was worked. On the upper and low parts of diffusion cell was closed with the metallic filters so that the water can intrude the cell and then wet the specimen. After the specimen was assumed has saturated with water, the upper part of soil sample specimen was dropped with 0.2 mL of $^{137}$Cs tracer and the diffusion process was begin. Diffusion experiments were done in 2, 4 and 6 months. The diffusion cell was opened, the soil sample specimens were cutted sliced with thickness 0.2 cm each. To measure their $\gamma$-activities of $^{137}$Cs in the sliced soil samples, we used a Canberra multichannel analyser (MCA) unit completed with HPGe detector 20% efficiency.

3. Results and Discussion

The profile of $^{137}$Cs concentration in the diffusion cell at the condition of dry density 1.41 g.cm$^{-3}$ were shown in Figures 5, 6 and 7. At Figure 5 with diffusion time was 2 months showed that most of $^{137}$Cs concentration accumulated at the surface or the area of near surface parts of soil sample specimen with intrusion distance of $^{137}$Cs was 1.4 cm. At the beginning of contact between $^{137}$Cs with soil sample, radiocesium gathered in the area near the surface of the soil sample in the diffusion cell, as seen at Figure 5. At that time there was an interaction of $^{137}$Cs with soil sample started.

When the diffusion time was added from 2 months to 4 months, the concentration of $^{137}$Cs accumulated at the near surface area started to distribute to the lower parts of soil sample and to flow to the further point with the distance range becomes 1.6 cm, see Figure 6. Most of the absorbed radiocesium was slowly released from the soil sample because the bonding that occurs between $^{137}$Cs and soil sample is only a Van der Walls bond that is not quite strong. The existence of the gravity force has made the radiocesium present in the soil sample goes deeper down to the bottom of the diffusion cell.

The distribution of $^{137}$Cs can reach the lowest part of soil sample with lower concentration of $^{137}$Cs however distance range of radiocesium more deeper than previous or shorter diffusion time, the result can be shown in Figure 7.
Figure 5. Concentration of $^{137}$Cs in soil samples at the diffusion cell as a function of squared distance, diffusion time was 2 months.

Figure 6. Concentration of $^{137}$Cs in soil samples at the diffusion cell as a function of squared distance, diffusion time was 4 months.
To obtain the diffusion coefficient (Da) of $^{137}$Cs in a diffusion cell, the equation (3) was applied. Da was obtained by plotted between the calculated result of log C with squared distance ($x^2$) of the center part of sliced soil samples. By using linear quadratic equation $y = ax + b$

Where $a = -\frac{1}{4Da t}$ and the the diffusion coefficient/Da becomes $D_a = -\frac{1}{4}$. The Da results was resumed in Table 1, and was shown in Figure 8.

The results of the experiments that have been performed can be resumed in Table 1. Increasing the time for diffusion causes the value of Da on the soil sample to become smaller, due to the large activity of $^{137}$Cs has been distributed to the deeper part of the soil sample layer from the high concentration to the lower concentration. So it will be found that the range of diffusion of $^{137}$Cs be getting deeper on the diffusion cell. The diffusion process occurs as long as gradient exists and becomes the major factor in mass transport in a materials even in a very low permeability condition of geological materials.

Table 1. The values of Da as a function of diffusion time

| No. | $t_{diffusion}$ (months) | Da $(m^2.s^{-1})$ |
|-----|--------------------------|-------------------|
| 1.  | 2                        | 3.58 E-12         |
| 2.  | 4                        | 3.28 E-12         |
| 3.  | 6                        | 1.76 E-12         |

$y=-0.5 E-13x + 5. E-12$ $R^2=0.869$

When the results are compared with other experiment results as has been done by [9-13], seen that the obtained value of Da has given a different value than the references, see Table 2. This is due to the differences in the use of experiment medium and also the diffusion time that is done. As a comparison of the results of the study, the most widely used material was bentonite [9-11] in addition to soil [12] and clay [13], the diffusion time used ranged from 4 days [11] to 17 years [12]. Bentonite has the smaller Da values although it uses in a shorter contact time, so bentonite material is often used as a buffer material in the radioactive waste disposal facility.
Table 2. Comparison of Da value of the experiment results

| No. | Da (m$^2$.s$^{-1}$) | Ref. |
|-----|------------------|------|
| 1.  | 1.76 x 10$^{-12}$ | This experiment |
| 2.  | 1.02 x 10$^{-11}$ | [9] |
| 3.  | 5.21 x 10$^{-12}$ | [10] |
| 4.  | 5.38 x 10$^{-11}$ | [11] |
| 5.  | 1.14 x 10$^{-12}$ | [12] |
| 6.  | 6.36 x 10$^{-10}$ | [13] |

Figure 8. Diffusion coefficient of $^{137}$Cs at diffusion cell as function of diffusion time.

The longer the diffusion experiment runs will decrease the Da value of radionuclide in the samples. The Da value of radionuclides will be affected by the initial concentration of radionuclides, density and also the ability of the radionuclides sorption by the medium [14,15].

Smaller of Da value on a medium will cause the transfer of radionuclides in the media is also getting slower. The expected low conditions of Da value occur in soils at the unsaturated zone could control the possibility of radionuclide spreading from disposal facility to the saturated or aquifer regions. Because groundwater located in the saturated zone is a highly effective medium for dispersion of radionuclides from the facilities to the environment.

4. Conclusions
The determination of diffusion coefficient of $^{137}$Cs at unsaturated zone of DH-2 site soil of SNC under d=1.41 g.cm$^{-3}$ condition has been performed, and the lowest of Da value was obtained after the diffusion experiment was conducted for 6 months with the Da value being 1.76 x 10$^{-12}$ m$^2$.s$^{-1}$. With increasing the diffusion time on a solid medium will cause the decrease in the value of Da.

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