Numerical Model for Leaching and Transporting Behavior of Radiocesium in MSW Landfill

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Abstract: This paper presents the numerical simulation model for radiocesium leaching and transporting behavior in municipal solid waste (MSW) landfill and discusses on the design for the required geometry and properties of the impermeable final cover and the soil sorption layer, which work for containment of hazardous waste such as radiocesium-contaminated MSW generated by Fukushima Daiichi nuclear disaster. The leaching behavior of the radiocesium from the waste was formulated based on the results of serial batch leaching tests, and the soil sorption was formulated using the nonequilibrium liner kinetics. In consideration of the leaching and soil sorption phenomena of radiocesium, COMSOL can solve the advection-dispersion governing equation predict the fate of the radiocesium that was leached from the landfilled radiocesium-contaminated MSW.

Keywords: Landfill Design, Leaching Behavior, Transport in Porous Media, Radioactive Half-life, Soil Sorption, Radiocesium

1. Introduction

On 11 March 2011, the Great East Japan Earthquake and subsequent tsunami occurred. They attacked Fukushima Daiichi Nuclear Power Station, resulting in loss of an electrical power for the cooling system of reactors and then the meltdown of nuclear fuel rods. The explosion of hydrogen gas in the electrical generation reactors caused the spread of the radioactive substances, mainly radiocesium, into the atmosphere. The radiocesium in the atmosphere fell down with rainfall, so that it contaminated all the things on the ground of soil surface, trees, branches and leaves, roads, concrete structure, farm products, sewage sludge, and so on. Because their waste and incineration ash include the radiocesium, their final disposal in landfill sites has to be carefully designed to prevent the radiocesium from leaking to the outside.

COMSOL MP v4.3 was used to numerically investigate the effects of the landfilled waste on the of the radiocesium concentration leachate from the landfill site and to propose the landfill design such as (1) acceptant concentration of the radiocesium included in the landfilled waste (2) permeability and geometry of soil sorption layer underlying the landfilled waste layer to adsorb the radiocesium leaching from the waste, (3) and permeability and geometry of impermeable final cover on the landfilled waste layer to prevent the rainfall from infiltrating the waste and leaching the radiocesium, to minimize the concentration of the leachate. Figure 1 illustrates the proposal for landfilling the waste contaminated with the radiocesium. This paper focuses on how to landfill the incineration fly ash contaminated with the radiocesium because the radiocesium is concentrated to the fly ash in the incineration process and the radiocesium in the fly ash can be easily leaching to water.

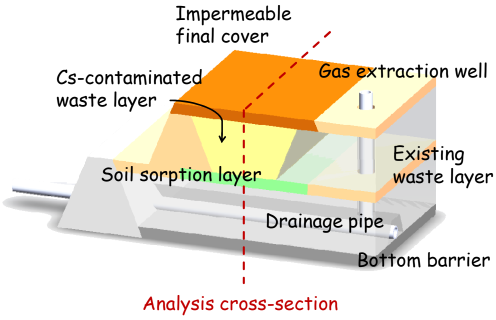

Figure 1. Proposal for landfilling waste contaminated with radiocesium

2. Leaching and sorption characteristics of radiocesium

Numerical analysis is effective to predict the radiocesium leaching and transporting behavior in the landfill sites and to design the required geometry and properties for the impermeable final cover and the soil sorption layer against the contaminated waste with a certain concentration.

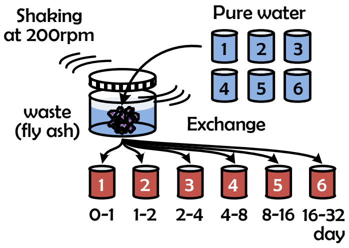

Figure 2. Serial batch leaching test

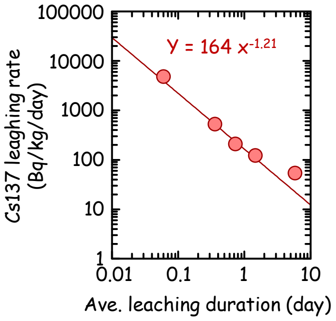

Figure 3. Radiocesium leaching rate from fly ash

First, the leaching behavior and soil sorption characteristics of radiocesium were investigated and formulated.

The leaching behavior of the incineration fly ash was investigated according to the serial batch leaching test as shown in Fig.2. The fly ash of was mixed with the pure water to be solid/liquid ratio = 10 mL/g. They were silently stirred with the propeller of 200 rpm. The water in the vessel exchanged new pure water on the 1st, 2nd, 4th, 8th, 16th, 32th day after the stirring started. The radiocesium concentrations in the exchanged water samples were measured by germanium semiconductor detector. From the measured concentrations, the radiocesium leaching rate was calculated as

$$J_i = \frac{c_i}{t_i - t_{i-1}} \frac{L}{S} \tag{1}$$

$$\bar{t}_i = \left(\frac{\sqrt{t_i} + \sqrt{t_{i-1}}}{2} \right)^2 \tag{2}$$

where, t_i = elapsed time (day), c_i = i-th concentration (Bq/L), J_i = i-th leaching rate (Bq/kg/day), L = liquid volume (L), S = sample mass (kg), and i = index of the fraction. The obtained leaching rates, J_i, were related to the average elapsed time, $\bar{t}_i$, in

$$J = K\bar{t}^{-a} \tag{3}$$

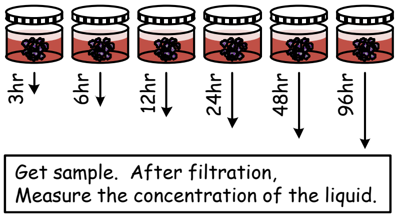

Figure 4. Parallel batch soil sorption test

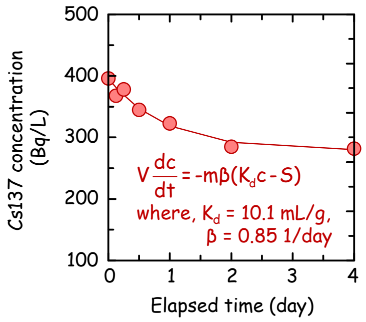

Figure 5. Radiocesium sorption capacity and reaction rate for decomposed granite soil (electric conductivity of used leachate 4200 mS/m)

where, K = initial leaching rate (Bq/kg/day) and a = leaching parameter. Figure 3 shows the leaching rate of the radiocesium 137 from the fly ash. The leaching behavior can be formulated in a time-dependent power function.

Soil sorption layer is effective to retard the diffusion of radiocesium leached from the waste and to decrease the concentration by radioactive decay during the retardation. In order to evaluate the soil soprtion ability, the batch sorption tests were performed as shown in Fig.4. The granite soil of 50 g was mixed with the waste leachate of 200 mL (solid/liquid ratio = 4 mL/g) in the bottle. The same six bottles were prepared. Each bottle was used for measuring the concentration of the radiocesium on 3hr, 6hr, 12hr, 24hr, 48hr, 96hr after the tests started. The relationship between the measured concentration and the elapsed time was fitted to the following nonequilibrium liner sorption model:

$$V\frac{dc}{dt} = -m\beta\left(K_d c - S\right) \tag{4}$$

$$\frac{dS}{dt} = \beta\left(K_d c - S\right) \tag{5}$$

where, m = soil sample mass (kg), V = leachate volume (L), K_d = distribution coefficient (mL/g), c = liquid phase concentration (Bq/L), S = solid phase concentration (Bq/kg), and β = equilibrium reaction rate (1/day). Figure 5 shows the change of the liquid concentration with the elapsed time. The decreased concentration due to soil sorption can be modeled with Equations (4) and (5).

3. Numerical Simulation

3.1 Governing Equations

When the landfilled waste layer is assumed isotropic porous media, the volumetric velocity of the water fluid flow is given by Darcy's law:

$$u_w = -\frac{k_{rw}K}{\eta_w}\left(\nabla p_w + \rho_w g \nabla z\right) \qquad (6)$$

where, u_w = volumetric gas velocity (m/s), k_{rw} = relative hydraulic conductivity, K = intrinsic permeability (m^2), η_w = water viscosity (Pa s), p_w = water pressure (Pa), ρ_w = water density (kg/m^3), and g = gravity acceleration (= 9.81 m/s^2). Thus, governing flow equation is as follows:

$$\frac{\partial(\rho_w\theta_w)}{\partial t} = \nabla\cdot\left[-\rho_w\frac{k_{rw}K}{\eta_w}\left(\nabla p_w + \rho_w g\nabla z\right)\right] \qquad (7)$$

here, θ_w = volumetric water content.

Transport of diluted chemical components such as the radiocesium is formulated as:

$$\frac{\partial(c_w\theta_w)}{\partial t} + \nabla\cdot\left(-\theta_w D\nabla c_w + u_w c_w\right) = -\theta_w\lambda c_w + R \qquad (8)$$

here, c_w = concentration in water phase (Bq/m^3), D = dispersion coefficient (m^2/s), λ = first-order ratioactive degradation rate (1/s), and R = source and sink term ($Bq/m^3/s$). The source/sink term, R, consists of the radiocesium leaching rate from the waste and the radiocesium nonequilibrium sorption/desorption reaction to soil as

$$R = \rho_d K t^a - \rho_d\frac{\partial S}{\partial t} \qquad (9)$$

where, ρ_d = dry bulk density of waste (kg/m^3), K and a = leaching parameters evaluated from the serial batch leaching test (see Fig.3), t = elapsed time (s), and S = concentration in solid phase (Bq/kg). Finally, mass balance equation in solid phase is formulated as:

$$\frac{\partial S}{\partial t} = \beta\left(K_d c - S\right) - \lambda S \qquad (10)$$

where, K_d and β = sorption parameters evaluated from the parallel batch soil sorption tests as shown in Fig.5.

3.2 One-dimensional simulation

In order to evaluate the effect of the rainfall on the radiocesium leaching concentration, the one-dimensional simulation was conducted by parametrically changing the rainfall intensity. Figure 6 shows analysis domain. The waste layer had a 3m height and the soil sorption layer had a 0.5m height. The fly ash in the waste layer had the radiocesium leaching concentration of 47 Bq/L (testing conditions were liquid/solid ratio = 10 and testing duration = 6 hour. The distribution coefficient of the soil sorption layer was 10 mL/g.

Figure 7 shows the results of the numerical simulations. The concentration of the leachate passing through the soil sorption layer depends on the rainfall intensity. The peak concentration of the leachate decreased as the rainfall intensity decreased, because the decreased flow velocity enlarged the retention time and the radiocesium can be degraded due to the radioactive decay effect. Thus, it was conclude that the design of the barrier performance of the impermeable final cover, which was installed on the radiocesium-contaminated waste, was important to minimize the radiocesium concentration of the leachate.

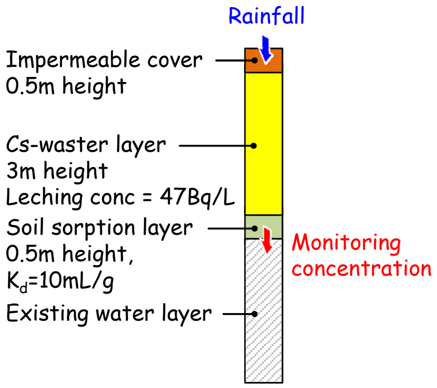

Figure 6. One-dimensional analysis domain

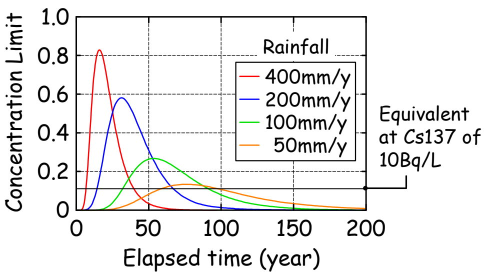

Figure 7. Concentration profiles of leachate passing through soil sorption layer

Table 1 Analysis conditions for two-dimensional simulation

	Unit	Existing waste	Cs-cont. waste	Cover soil	Soil sorp. layer	Imp. final cover
Permeability	m²	10^{-12}				10^{-16}
Porosity	---	0.3				
Dry bulk density	kg/m³	1200		1800		
VG parameter, a	1/m	3.01				
VG parameter, n	---	1.26				
Distribution coef.	mL/g	0			10	0
Cs leaching rate	Bq/kg/day	0	$82(t)^{-0.5}$	0		

3.2 Two-dimensional simulation

Two-demensional simulation was conducted in order to investigate the effects of the hydraulic conductivity of the impermeable final cover and the geometry of the radiocesium-contaminated waste layer on the concentration of the leachate. Figure 8 shows the analysis domain, and it was the same cross-section in Fig.1. The hydraulic conductivity of the impermeable final cover was set at 10^{-5} or 10^{-7} cm/s, and other parameters such as the leaching parameters were the same as those shown in previous section.

Figure 9 shows the concentration distribution of the radiocesium 137 in the analysis domain. When the impermeable final cover to prevent infiltration of the rainfall had a low hydraulic conductivity, the rainfall cannot infiltrate the radiocesium-contaminated waste layer. Hence, the radiocesium can remain in the waste layer so that the concentration of the radiocesium can be decreased due to the radioactive degradation.

Figure 9 shows the effects of the soil sorption layer and the impermeable final cover on the concentration of the leachate. Compared with the no countermeasure (without soil sorption layer and impermeable final cover), soil sorption layer and impermeable final cover can work for decreasing the peak concentration. In particular, when the landfill has both the soil sorption layer and the impermeable final cover, the radiocesium was hardly included in the leachate.

4. Conclusions

COMSOL Multiphysics simulations were useful in public explanation for the effects of the soil sorption layer and the impermeable final cover on the decrease of the radiocesium emission and their importance. In addition, the parametric sweep can be used for determining their permeability and geometry conditions.

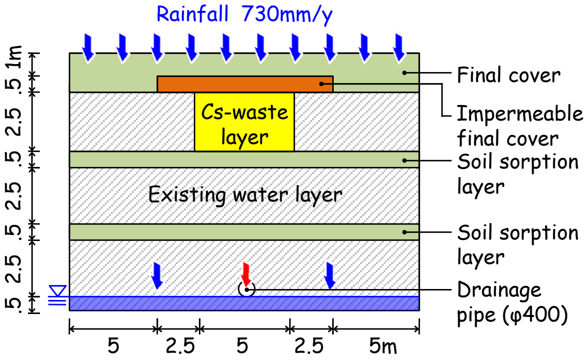

Figure 8. Two-dimensional analysis domain

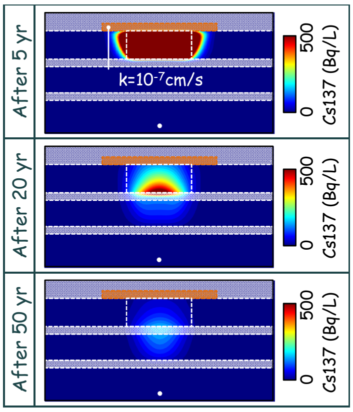

Figure 9. Radiocesium concentration for hydraulic conductivity of 10^{-7} cm/s in impermeable final cover.

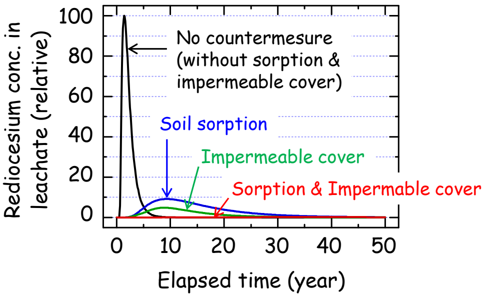

Figure 10. Effects of impermeable final cover and soil sorption layer on leachate concentration