A Study on the Performance of Flocculating agent for Radioactively Contaminated Soil by Soil Washing Process

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Abstract

Radioactive substances that are discharged in the process of nuclear power plant Decommissioning or facility maintenance cause soil pollution. Such soil pollution requires decontamination in order to ensure people's residence and reuse of land as an industrial site. Among the various soil restoration technologies, this paper selected soil washing technology, which has actual example of being used on soils polluted by radioactive substances in and out of the country. Soil washing process uses cleansing agent to weaken the surface tension of soil and Cs in order to separate soil and Cs. In order to enhance efficiency of process by reusing the cleansing water in soil washing process, Sedimentation agent was used in the cleansing water to perform the experiment for studying the performance of removing micro-soil and Cs. Also, Cs removal experiment results using ICP-OES and rate of combination between Cs and soil using Visual MINTEQ Code were found out in order to compare Cs and micro-soil removal performance in cleansing water.

1. Introduction

Radioactive substances are discharged in the process of nuclear power plant Decommissioning and facility maintenance. Major substances of such discharged radioactive substances where decontamination by soil pollution ought to be considered include Cs-137 and I-131.

Among these, Cs-137 should be considered with priority, because Cs-139 is water-soluble and highly toxic, producing harmful effects when it enters the body through breathing or consumption. Technology that has actually been used to remove Cs includes soil washing, soil cleansing, and current electricity, and the technology which decontaminates a wide range of soil in a short period of time and shows a relatively high decontamination efficiency is the soil washing process [1].

Soil washing process uses appropriate cleansing agent to weaken the surface tension of soil and Cs in order to separate the soil and Cs. The major factor that determines decontamination efficiency is the selection of cleansing water. While the decontamination process which uses acidic cleansing water weakens surface tension greater than the one that uses water, therefore separating Cs and soil with high efficiency, it produces much acid effluent. Using water instead of acidic cleansing water, which produces great amount of effluent, and reusing cleansing water using sedimentation agent may ensure a more economical and efficient process. In order to enhance the efficiency of soil washing process, which uses water as the cleansing water, by reusing the cleansing water, this study used sedimentation agent to perform the experiment for testing the removal performance of micro-soil and Cs that are included in the cleansing water. Also, in order to compare the experiment results through ICP-OES, it used Visual MINTEQ Code to compare the removal performance of Cs and micro-soil in cleansing water.

2. Soil Washing Process

2.1 Soil Remediation techniques

In general, soil restoration technology largely consists of Physicochemical, biological, and thermal process as shown in Table 1. Among the soil restoration technologies, biological process, where much time is required in case of pollution by radioactive substance, as well as thermal process, where the efficiency of removing radioactive substance is low, is not appropriate to be used. Therefore, soil restoration technologies for removing radioactive substances are Physicochemical processes with methods that can process heavy metals, such as soil washing, current electricity, and heap-leaching [2].

Classification	Techniques	Feature	
Physicochemical treatment	Soil-washing		
	Electro-kinetic	• Easy to use in combination with other processes	
	Soil-flushing	• Soil remediation time is generally short and efficiency is high	
	Heap-leaching		
Biological treatment	Biodegradation	• Efficiency in terms of heavy metals is	
	Land farming	generally low · Requires time equal to half-life of nuclear species to exhibit	
	Natural attenuation	decontamination efficiency for radioactive materials	
Thermal treatment	Thermal desorption	· High decontamination efficiency for	
	Vitrification	radioactive nuclear species but no reduction of waste	
	Pyrolysis	volume · Secondary waste such as ash, etc.	

Table 1. Soil remediation techniques

2.2 Soil washing techniques

Among Physicochemical soil remediation technologies, soil washing process shows high decontamination efficiency within a relatively short period of time, therefore having actual cases of being utilized in soil polluted by radioactive substances. Actual domestic case example includes the case of Korea Atomic Energy Research Institute (KAERI) for decontaminating the soil near TRIGA Mark-II&III that included Co-60 and Cs-137 [3]. Foreign case includes the case of Oak Ridge National Lab using soil washing for Cs-137 [4].

Soil washing process largely consists of Size Reduction Equipment, Screening Equipment, Separation Equipment, Cyclone Equipment, and Waste Water Treatment Equipment.

As shown above, a large quantity of cleansing water is required in order to remove Cs in soil. In the process, reusing the waste water from Waste Water Equalization Tank by circulating it without discharging it will lead to a more efficient soil washing process. For efficient reuse of cleansing water, sedimentation agent was put into the cleansing water to conduct the experiment on Cs and soil removal performance.

3. Experimental Method

To reuse the cleansing water for soil washing process, Sedimentation agent (J-AF, JeonTech Co.,Ltd.) was used to perform the experiment on micro-soil and Cs removal performance. In natural environment in general, Cs is strongly united with soil with small particle size. In order to create an environment where Cs is mostly adsorbed on soil with small particle size, the soil was separated using filter into approximately 38μ m, and the filtered soil was dried for 20 minutes using vacuum drier. Also, in order to ensure that the soil and Cs can sufficiently be united, 1g of filtered soil was prepared with Cs concentrations of 0.1, 1, 2, 5 and 10mmolee and then stirred at 250rpm for 24 hours. CsNO₃ was used as Cs specimen, temperature was 23° C, and pH was 6.5.

In the first experiment, Sedimentation agent was used to analyze maximum rate of removal and rate of removal by time. For the experiment, 0.1g of Sedimentation agent was put into soil including Cs that has been stirred for 24 hours, stirred for 5 minutes at 450rpm to extract supernatant, and filtered using 0.2μ m(Cellulose Nitrate Membrance Filter) in consideration of Cs particle size. Finally, Cs remaining in experiment solution was analyzed using ICP-OES (Parkin Elaer, US/Optima 2100).

Also, to study Cs removal performance of Sedimentation agent by time, supernatant of the experiment solution was extracted five times, by 1 minute each time, and filtered using 0.2ufilter. Then,

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Fig 1. Sediment removal performance for each time



Fig 2. Sediment removal performance for each concentration

10 mmole, 0.1g of Sedimentation agent was injected to test Cs removal performance as shown in Fig. 2.

4. Results and discussion

When using 0.1g of J-AF Sedimentation agent in the experiment, maximum Cs removal performance was approximately 88% at Cs concentration of 0.1mmolee, approximately 85.8% at concentration of 1mmolee, approximately 85% at concentration of 2mmolee, approximately 71% at concentration of 5mmolee, and approximately 67% at concentration of 10 mmole.

To review maximum Cs removal performance that was resulted from the experiment using ICP-OES, Visual MINTEQ Code, which is a chemical equilibrium code, was to derive the binding rate of Cs and soil, and turbidity was multiplied on the binding rate to compare the values. Turbidity is measured to study soil removal rate of Sedimentation agent.

Visual MINTEQ code is a chemical equilibrium model that was developed to calculate equilibrium, sorption, adsorption, etc. in an aqueous solution. It is a code that uses Visual Basic Interface based on MINTEQA2 software of USEPA (KTH, Jon Petter Gustafsson)[5].

To apply soil and Cs concentration on the Input value of Visual MINTEQ Code, replaceable cation and anion concentrations in soil were applied by converting them into units of ppm [6]. Binding of Cs and soil using Visual MINTEQ Code is shown in Table 2. The amount of replaceable cation and anion is shown in Table 3 [7]-[8].

	Cs concentration (mmole)						
	0.1	1	2	5	10		
Cs	8.433	8.605	8.801	9.431	10.633		
CsCl	47.877	47.946	48.014	48.172	48.271		
CsSO4	15.417	15.230	15.028	14.455	13.595		
CsNO	28.272	28.219	28.157	27.943	27.502		

Table 3.Concentrations of cations and anions in soil

Soluble cation and anion (cmolc/kg)							
K	Ca		L		Mg		
1		5.3		1.7			
Concentration of Anion(cmolc/kg)							
Cl		N0a	PO4		SO4		
4		7.1	4.8		7.7		

Also, turbidity (LAMotte 2020, Turbidity Meter) was measured to study soil removal rate in the experiment solution. Turbidity was conducted in the same conditions with those of the experiment using Sedimentation agent. Initial turbidity of muddy water was 47.2NTU. After injecting 0.1g of Sedimentation agent, the solution was stirred for 5 minutes, and then supernatant was collected and filtered using 0.2µm filter to measure turbidity. Turbidity after using Sedimentation agent was measured to be 4.39NTU, showing that Sedimentation agent removes soil with Cs by approximately 90%.

Fig 4 shows a comparison of the values from using ICP-OES with the values multiplying the soil removal rate of the sedimentation agent obtained from measuring turbidity (NTU) before and after adding the sedimentation agent by the binding rate between Cs and the soil obtained from the code.



Fig 3. Comparison of ICP-OES and Visual MINTEQ Code

The Cs removal performance of the sedimentation agent using ICP-OES and the product of the code value and turbidity indicated conservative trends at lower concentrations, but it was shown that values differed in the approximately 2 mmole or higher concentration levels. Based on this, it is expected that the Code and ICP values will exhibit sufficiently similar trends at concentrations of 2 mmole and higher as well if sufficient stirring time of 24 hours or more is allowed for the Cs and soil.

5. Conclusions

Methods for removing radioactive nuclide that is exposed to soil when decommissioning or repairing power plant have been applied in domestic and foreign cases. Soil washing process, which has a relatively short decontamination period, is most appropriate for use. Experiment result of Cs removal using Sedimentation agent at an environment that assumes to use soil washing process that uses water as the cleansing agent showed that maximum of approximately 90% of Cs was removed at Cs concentration of 0.1 mmole. This indicates that cyclical recycling of cleansing water is possible. This result shows that soil washing process can be operated in a very efficient and economical manner within a short period of time. Also, the value of Cs removal performance of Sedimentation agent using ICP-OES and the value of multiplying the binding rate of soil and Cs by soil removal rate of Sedimentation agent using Visual MINTEO Code showed a similar trend. If the actual data obtained from analyzing the soil component around the actual

NPPs and estimating the Cs and the amount of radioactive materials, which will be exposed to the soil, can be applied to the code, it is possible to obtain more accurate values in the future. And it is anticipated that the main output from the code as the main data will be used to predict the rate of soil remediation and to select a decontamination method satisfying the site release criteria.

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