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How to Remove the Radioactive Cesium in Contaminated Soil?

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1. Introduction

After the nuclear power plant accidents, a huge amount of radioactive material has deposited on the surface of the earth. Among the radioactivities, nuclides with very long half-lives are distributed over several layers of soil over a long period of time, and many researchers have reported that they affect the environment [1-2]. In particular, Kimiaki Saito et al. told that the effective doses evaluated for external and inhalation exposures due to radioactive cesium (Cs-134, Cs-137) were much higher than those due to other radionuclides were in the contaminated area of Fukushima [3].

On the other hand, efforts are being made to remove soil pollution as part of Fukushima reconstruction. It is confirmed that the work was started to put the surface soil into plastic bags and move to an intermediate storage area centering on Fukushima 1 power plant. The useful information provided by studies on the depth profile of radioactive cesium in contaminated soils could be used to estimate the rate of radioactive cesium downward movement [4]. In addition, a study on the evaluation of the air dose rate according to the vertical and horizontal distribution of radioactive cesium has confirmed that the removal of the topsoil is an effective decontamination method [5]. However, since the contaminated area due to the Fukushima accident is so vast that the amount of soil to be removed is enormous, and the cost of collecting and storing it is too much, it is necessary to consider a more efficient method.

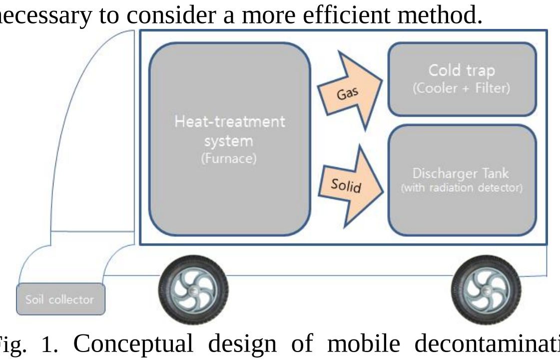

Fig. 1. Conceptual design of mobile decontamination system for radioactive soil.

If the soil is heat-treated and only cesium is vaporized and collected by the filter, there is an advantage that only a filter having a much smaller volume can be treated as waste, compared to the case where the entire contaminated soil is disposed of. In addition, it is expected that a simple mobile system composed of functional elements as shown in the Fig. 1 will be able to perform decontamination immediately in the field. However, JAEA's 2012 report says that cesium was not removed from the contaminated soil after heat-treatment [6]. In this study, the change of cesium content in soil samples was observed in various heat treatment conditions and the possibility of cesium removal by heat treatment was examined in soil.

2. Methods and Results

More efficient way using the physical and chemical properties of the radioactive element is proposed. The effectiveness of the proposed method is verified by the heat treatment experiment and mass spectroscopy.

In order to check the effect of heat treatment on cesium vaporization in soil, soil samples were obtained from the Jeju island and Cs_2CO_3 (99%, Sigma-Aldrich) was added to these samples. The cesium elements cannot be existed as the form of pure Cs in soil due to its high reactivity so that we assumed that Cs_2CO_3 is one of possible phase for Cs in soil. The amount of Cs_2CO_3 was varied from 1wt% to 4wt%. The samples with Cs_2CO_3 was annealed at 450, 550, 750, 1000, and 1100°C for 1 hour under Ar and air atmosphere.

The phase of the soil samples was identified using X-ray diffraction (XRD, SmartLab, Rigaku) with Cu Ka radiation. The volumetric phase fraction was analyzed by the Rietveld refinement method, implemented in Profex [7]. The composition was analyzed using inductively coupled plasma mass spectroscopy (ICP-MS, NexION 300D, PerkinElmer) and X-ray fluorescence (XRF, RIX 2100, Rigaku). The thermal stability was investigated by thermogravimetric analysis (TGA, TGA/DSC1/1600LF, Mettler Toledo).

The composition of the soil without any addition of Cs_2CO_3 and heat treatment, analyzed with XRF, is shown Table 1. The main elements in the soil were Fe, Si, ad Al. The composition of soil can be varied with places in which the soil was obtained. For examples, the soils obtained at forest of Lipowa Bugaj in Poland have Si (28.9 wt%), Al (8.9 wt%), and Fe (3.7 wt%) as main elements [8]. The large amount of Fe is a characteristic feature of the soil of Jeju island.

In order to confirm the content of the added Cs_2CO_3 in the soil, the composition of Cs was analyzed with ICP-MS, whose results are shown in Fig. 2. The contents of Cs is gradually increased with the nominal amount of Cs is increased, which means Cs is properly added to the soil by means of the Cs_2Co_3 compounds and ICP-MS can be applicable to detect Cs in the soils.

Table 1. Compositional analysis of soils obtained from Jeju island with XRF

Elements	Mass %	Elements	Mass %
Fe	36	Na	0.484
Si	30.8	Br	0.273
Al	20	Cl	0.123
Ti	4.62	Sn	0.114
K	2.07	Zr	0.109
Mg	1.22	Cr	0.0866
Ca	1.13	Zn	0.0725
P	1.09	Ni	0.0556
S	0.944	F	0.0325
Mn	0.647	Residues (Cu, Sr, Pb, Y, Ga, Nb)	0.1276

The Cs content in the soil sample after the heat treatment was investigated with ICP-MS. The amount of Cs is gradually decreased with the heat treatment (Fig. 3). The reduction of Cs is originated from the decomposition of Cs_2Co_3 to Cs_2O and volatile behavior of Cs_2O at high temperature. Even though the circumstance of Cs_2CO_3 and Cs_2O is quiet complicated, the vaporization of Cs is well processed in our soil samples.

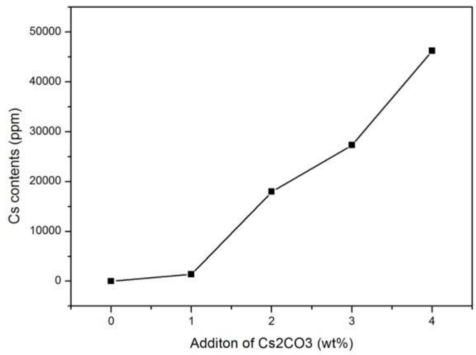

Fig. 2. Cs contents, analyzed by ICP-MS, of various Cs_2CO_3 added soil samples.

The difference between JAEA's 2012 report and ours could be originated from these aspects: 1) the compositional differences between the Jeju island and Japan, 2) inhomogeneous heat conduction and 3) atmosphere. As we showed already, the large content of Fe in the Jeju's soil is peculiar characteristics. Therefore, the study on the effect of composition of the soil will be needed in near future. In JAEA's 2012 report, torch flame was used to heat up soils. In this case, the heat cannot penetrate deep inside so that it will be difficult to make homogeneous reaction. The atmosphere during heating with torch is air. The lager partial pressure of CO_2 in air will reduce the reaction rate of Cs_2CO_3 decomposition. Therefore, we elucidate the reduction of Cs_2CO_3 through heat treatment in inert gas atmosphere, which will be applicable in reduction of soil's pollution contained with Cs.

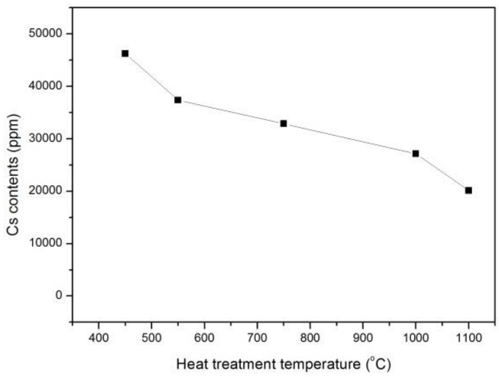

Fig. 3. Cs contents, analyzed by ICP-MS, of 4 wt.% Cs_2CO_3 added soil samples after heat treatment for 1hour under Ar atmosphere.

The results of phase analysis and thermogravimetric analysis due to heat treatment are still under analysis. We expect to have a chance to report again in the future.

3. Conclusions

It was confirmed that cesium in soil was removed by heat treatment. The relationship between soil properties and heat treatment effects still needs to be interpreted.

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