

# STUDY ON REMOVAL OF CONTAMINATED SOIL ON FOREST SLOPE FOCUSING ON DIFFERENCE IN DENSITY OF SUBSURFACE GROUND

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**ABSTRACT:** A large amount of radioactive cesium was released from the Fukushima Daiichi nuclear power station. Decontamination in living area has been almost completed for seven years. However, decontamination has not progressed much in forests where people do not enter. The decontamination of soil on forest slope cannot be done using heavy machinery unlike that on flatland. The effective decontamination method for the forest slope is desired. Therefore, in this study, the collecting method of slope surface soil through flushing water and the soil separation method focusing on the difference in the density of minerals were proposed and examined. In the proposed method, it is expected to reduce the volume of contaminated soil through the separation of the contaminated part and non-contaminated one. In this study, the laboratory test is conducted to evaluate the usefulness of the proposed method. In the test, a device of simulating the slope and a device to flush the soil by flowing water is produced. When an actual soil is utilized for the experiment, it is difficult to ensure the experimental repeatability due to soil variability. Therefore, simulated soil consists of Toyoura standard sand and vermiculite. To simulate soil contaminated with cesium, cesium standard solution is added into the simulated soil. The soil is set on the slope, and the test is conducted to flush and collect the soil by running water from upside to downside. As a result, it is clarified that only soil containing cesium can be removed by separating the soil after flushing.

*Keywords: Radioactive cesium, Forest slope, Vermiculite, Contaminated soil*

## 1. INTRODUCTION

The Great East Japan Earthquake, which is the most powerful earthquake on record, occurred in Sanriku offshore on March 11, 2011. The Fukushima Daiichi nuclear energy plant of the Tokyo Electric Power Company (1F) was hit by the great Tsunami several times after an hour of the earthquake. The power facilities were widely destroyed, and the basement room and the shaft were inundated with the Tsunami. The major quantity of radioactive materials was discharged from the 1F. The radioactive materials are spread to the atmosphere, which is fallen down mountain, river and marine by rain breeze. Also, the radioactive materials that have fallen into the mountains are firmly bonded to the clay minerals in the soil as in [1], [2]. Especially, the cesium-137 has been extensively detected in Fukushima prefecture because the cesium-137 has been vigorously discharged and a long half period. The radioactive cesium has been decontaminated by removing the topsoil because it has settled in the surface layer of the soil after as in [3] being absorbed by atmospheric dust as in [4]. The decontamination around the living environment of the resident is almost completed due to a hard decontamination

work since the accident. On the other hand, there are places where the decontamination is completed and decontamination is uncompleted in forests away from the living environment of residents. The completion ratio of decontamination of forests in the area where the country implements the decontamination is shown in Table 1. Also, from this table, there is a difference in the completion ratio of the decontamination depending on geographical features. Some forests in the area such as Iitate village and Kawamata town are scarcely decontaminated. Although the air dose rate of forest decreases year by year due to the decontamination work and half-life, it is still higher than that in the living area. The decontamination in forests is difficult because it is hard to enter heavy machinery to remove the topsoil. The decontamination of contaminated soils on flat grounds is primarily done by removing the topsoil. Furthermore, on forest slopes, careful decontamination work is required as excessive removal of topsoil can cause landslides.

For radioactive cesium in the forest soil, a removal method using wood chips is devised by Kaneko et al. at Yokohama National University as in [5]. The method utilizes the property that fungi absorb radioactive cesium. The wood chip is placed in the bag and radioactive cesium is adsorbed into

the cellulose body contained in the wood chip. After that, the decontamination is completed by collecting the bag. This method is inexpensive. However, it takes time because it can be removed only by the power of fungi. Alternatively, the previous studies are conducted by the faculty of engineering, Ibaraki University in order to control the moving of cesium on the forestry slope, and it has two features as in [6]. One is that the bentonite is spread onto the leaf mold in the forest so as to control the resorption of the radioactive cesium to plants. The other is that the moving of the bentonite including the radioactive cesium is controlled by polyion-complex (PIC) of excess positive charge and negative charge. The radioactive cesium in the air and solution is adsorbed into the bentonite. The resorption to the plant is controlled by the absorption performance of the bentonite. Alternatively, the forest ecosystem is not destroyed because the elements such as the flow of rainwater are used in this method. Furthermore, the PIC is harmless because the raw material of PIC is commonly used as the thickener of ice-creams as in [7]. The moving of cesium is controlled, but the contaminated soil is not reduced in this method. Besides, it takes time to complete the work due to the use of rainfall. Based on these points, in the previous study, the simple decontamination method is proposed in order to decontaminate the contaminated soil on the slope by flowing water as in [8]. From this study, it was clarified that it was possible to remove about 50% of cesium by running water from simulated contaminated soil and stirring after recovery of running water. On the other hand, it was mentioned as a problem that the variability occurred in the flowing water time and the flowing water amount in the produced equipment. Thus, in this study, the reproducibility of the test is improved by improving the equipment. Then, the method of recovering only soil containing cesium is proposed by using the difference in the specific gravity of the soil after collection. In this method, an only clay mineral that specifically adsorbs cesium such as vermiculite is recovered from mixed sand and separated. Furthermore, other cesium is recovered

by washing thorough stirring, which leads to a volume reduction of contaminated soil.

## 2. CESIUM IN THE SOIL

A large number of radioactive materials was released by the accident in the IF. The radioactive cesium has harmful long-term effects due to its long half-life. The volume of spreading strontium-90 is comparatively small, despite its long half-life. The cesium exists as the monovalent cation in the soil, and it is absorbed into the soil particle having a negative charge. Especially, the cesium is selectively absorbed into the small clay particle. The cesium is adhered to the soil due to the 2:1 types of lamellar silicate. When the part of silicon or aluminum substitute for another element having a few positive charges in the silicon tetrahedron sheet or aluminum octahedron sheet, the negative charge is expressed with the shortage of positive charge in the sheet. Moreover, the cation is absorbed in order to neutralize the intercalation. The charge in this intercalation shows a high selectivity for the cesium ion. That is because the six-membered ring is nearly equal to the ion radius of the cesium ion in the basal seat of the silicon tetrahedron sheet. Alternatively, the bonding strength with the six-membered ring becomes larger as the hydration energy is smaller, and the bonding strength of cesium ion is the highest. Generally, the six-membered ring is occupied by the potassium ion because the abundance of potassium ion predominantly is major in the soil (see Fig. 1 of top

Table 1 Decontaminated situation of forest in town and village

| Cities           | Decontamination completion ratio (%) |
|------------------|--------------------------------------|
| Tamura city      | 100                                  |
| Kawauchi village | 74                                   |
| Iitate village   | 3                                    |
| Kawamata town    | 12                                   |
| Katsurao village | 67                                   |
| Okuma town       | 44                                   |

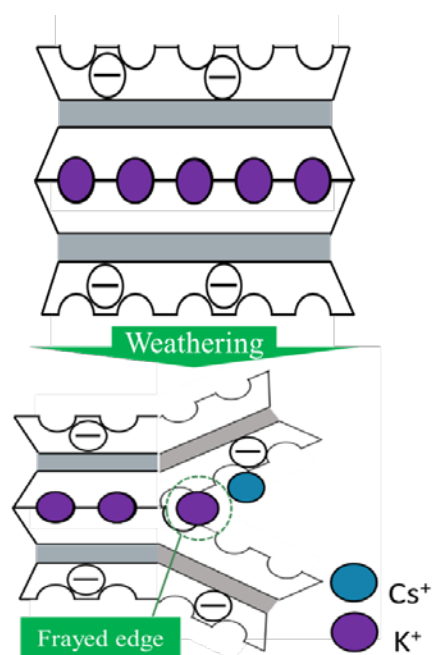


Fig. 1 Top part of 2:1 types of lamellar silicate and bottom part of the Frayed edge

part). The terminal of the layer in the clay mineral is swelled by the disintegration, and the potassium ion is discharged. Thus, the empty space that another cation gains entry is formed. This space is called the frayed edge (see Fig. 1 of bottom part), and the extraction of cesium ion from the frayed edge is very difficult. It is almost impossible to extract the cesium in the frayed edge only by flowing water. Thus, in this study, it is considered that the contaminated surface soil can be collected with the flowing water by utilizing the strong fixation.

### 3. METHOD

The simulated soil used in the test and the test method are explained, respectively. Alternatively, in this study, all of the tests are conducted three times under the same conditions to ensure the repeatability of the tests.

#### 3.1 Assumed Contaminated Soil

In this study, the Toyoura standard sand, vermiculite or their mixed sand is used as the assumed contaminated soil to which the cesium is added. The Toyoura standard sand consists of uniform grain size. The vermiculite contains the silicate mineral that mainly consists of the oxidized silicon, magnesia and aluminum oxide. The vermiculite has a high adsorption performance for cesium as in [9], and the mechanism of absorption has been clarified as in [10]. The vermiculite is widely distributed to Fukushima soil, and the vermiculite constitutes barriers to the decontamination of cesium in a soil as in [11]. Based on the past study as in [8], 9.5 g of the standard sand and 0.5 g of vermiculite is mixed in order to make the assumed contaminated soil.

#### 3.2 Test and Analysis Method

In order to imitate the real behavior of cesium in the soil slope, the slope device is produced (see Fig. 2). The slope device consists of two equipment: one is the slope section and another is the flowing water section. A 3D making device is used to produce the slope section (20.0 cm x 20.0 cm x 6.5 cm). The slit is set at the bottom of the slope section in order to collect the water and soil which contain cesium. Although details will be described below, the assumed soil falls into this slit by flushing water from the upper part of the slope of the equipment. Also, the slope angle is set to about 9 degrees to avoid the flow disruption. The flowing water section is made by the polyvinyl chloride (PVC) pipe having the spiral holes. The rotating shaft is set on the top of the slope, and the PVC pipe and the rotating shaft are connected. The PVC pipe is rolled

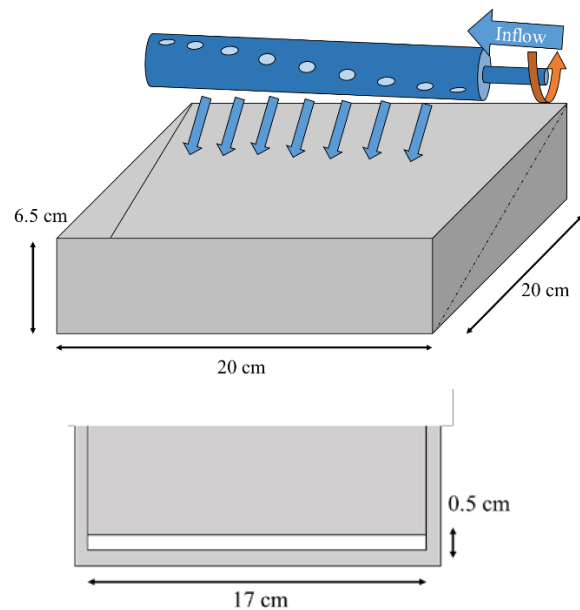


Fig. 2 Slope device

at 10 rpm, and the water is evenly flowed out from each hole by rolling the PVC pipe.

The test is divided into two steps. In the first step, the mixed sand containing cesium on the slope is recovered by running water, and the cesium is separated from soil by stirring. Specifically, 1 ml of a cesium chloride solution (about 3.3 ppm of initial concentration) is added to the mixed sand and the curing of mixed sand is conducted. It is known that the time required for adsorption of cesium to clay minerals is long. Also, a long period has elapsed since radioactive cesium deposited in the soil of Fukushima. Based on these points, a curing period is established to investigate the influence of the passage of time on the absorption of cesium in the soil. The curing period is 0, 7, 14 and 28 days. After curing, the mixed sand is set on the slope of the equipment. The assumed soil is collected by flushing water from the rolling pipe at the upper part of the slope. The water and assumed contaminated soil are collected from the slit by flushing water. The cesium concentration in the supernatant of the solution is analyzed by an AAS (Atomic Absorption Spectrometer). In the second step, the recovered the assumed contaminated soil is classified by sieving. From some trials of sieving at an early stage, it is found that the sieving of the sand is difficult under the moisture condition of the sand. Therefore, the assumed contaminated soil is dried for 24 hours in an oven dryer set at 110 °C. The using sieve size is 425, 250 and 106  $\mu\text{m}$ . After classification, the assumed contaminated soil remaining in each sieve is stirred, and the cesium concentration is analyzed by the AAS (Atomic Absorption Spectrometer) to calculate the extraction ratio for soil that remained

in each sieve. As the screwing method, 200.0 mL of pure water is poured into the collected soil, and the sample is screwed with 300 rpm for 10 minutes by the stirrer. The calculation formula of the extraction ratio is shown below.

$$\text{Extraction ratio (\%)} = \frac{\text{extracted amount (mg)}}{\text{added amount (mg)}} \times 100 \quad (1)$$

#### 4. TEST RESULTS

The results of the test in each step are shown and discussed, respectively. Each result is shown through the graph. The value in the graph is the average one of the test of the three times.

##### 4.1 Test Result of the First Step

In the first step, the assumed contaminated soil is recovered by flushing water, and the concentration of cesium in the supernatant of the recovered sample is analyzed by AAS. The results are shown in Fig. 3. The extraction ratio of cesium and the curing period of cesium are shown in the vertical and horizontal axes, respectively. In the specimens without the curing period, the extraction ratio of cesium is about 7%, and over 90% of cesium remained in the assumed mixed soil. The extraction ratio of cesium is low in the case of setting on curing period of cesium compared to the case of the specimen without curing period. The extraction ratio hardly changes even if the curing period is prolonged. It is found that the extracting of cesium from contaminated soil in a real situation is difficult by only flushing water regardless of the length of the curing period.

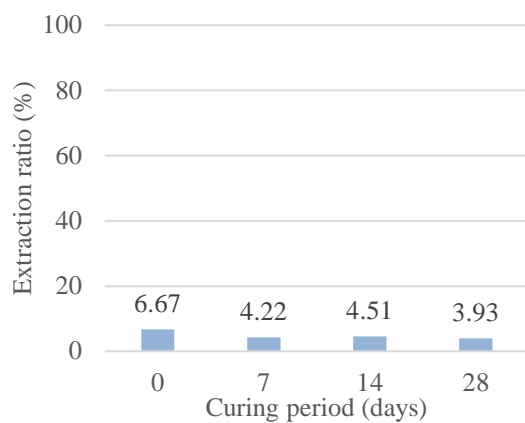


Fig. 3 Extraction ratio of cesium after flushing water

##### 4.2 Test Result of the Second Step

In the second step, the recovered soil in the first step is dried, and classified by sieving. The amount

of recovered soil is measured. After measuring, the sieved soil is washed out by stirring, and the concentration of cesium in the solution which is used during the stirring is analyzed by the AAS.

First, the mass of the assumed soil remaining in each sieve is shown in Table 2. The mass of the assumed soil before the test is 10 g, which means that almost all assumed soil is recovered by flushing water. It is suggested that the flushing water is effective for collecting the contaminated soil on the slope. Alternatively, all of the soil remaining in 425 μm of the sieve are the vermiculite because all standard sand pass through 425 μm of the sieve.

Secondly, the extraction ratio of cesium extracted by stirring is shown in Fig. 4. The extraction ratio of cesium and the curing period of cesium are shown in the vertical and horizontal axes, respectively. The lower the extraction ratio is, the longer the curing period is getting. It is considered that the cesium is adsorbed into the frayed edge in vermiculite by curing as the reason. Moreover, the cesium tends to be extracted easily from the soil that remained in 106 μm of the sieve. For this reason, it is presumed that most of the soil remaining in 106 μm of the sieve is the standard sand. From these facts, it is suggested that the concentration of cesium can be reduced by screwing the soil except

Table 2 Mass of soil remaining in each sieve

| Sieve mesh (μm) | Curing period (days) |      |      |      |
|-----------------|----------------------|------|------|------|
|                 | 0                    | 7    | 14   | 28   |
| 425             | 0.46                 | 0.45 | 0.44 | 0.48 |
| 250             | 1.09                 | 1.35 | 1.38 | 1.93 |
| 106             | 8.40                 | 8.13 | 8.13 | 7.53 |
| Bottom          | 0.03                 | 0.03 | 0.03 | 0.02 |
| Total           | 9.99                 | 9.97 | 9.98 | 9.96 |

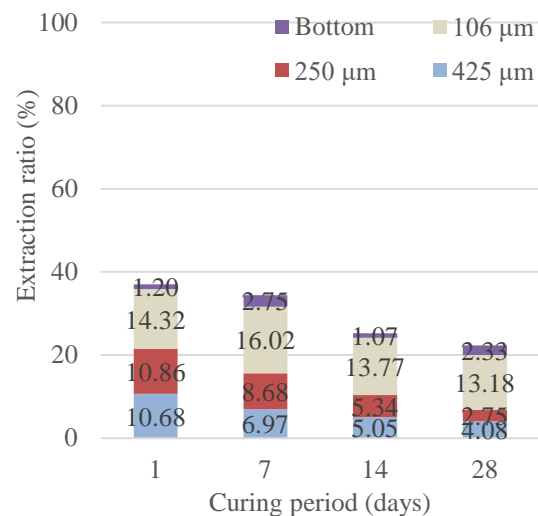


Fig. 4 Extraction ratio of cesium after sieving and screwing

frayed edges such as the standard sand. Also, the amount of contaminated soil may be reduced by separating the vermiculite containing a lot of cesium from the soil.

## 5. CONCLUSIONS

In this study, the test equipment assuming the forest slope is made and the recovering test of the assumed contaminated soil by flushing water is conducted in order to establish a new decontamination method for the soil of the forest slope where the decontamination work is hard. The standard sand and the vermiculite are mixed to model the assumed contaminated soil. The contaminated part of the soil can be reduced by separating and recovering the soil because their adsorption property of minerals involved in the soil for cesium is a great difference. Therefore, the recovered contaminated soil is classified according to the minerals by the mesh control with a sieve. After classification, washing out by stirring is attempted to reduce the concentration of cesium in the assumed contaminated soil by separating the main part in which cesium is mainly involved. As a result of the test, the assumed soil is recovered with only flushing water. Furthermore, the standard sand and the vermiculite are separated by the mesh control with a sieve. In other words, it is suggested that only vermiculite adsorbing cesium strongly should be recovered to reduce the contaminated soil. The extracting cesium from the separated standard sand with stirring may also lead to the reduction of the concentration of cesium in the contaminated soil. From the above results, it is considered that the proposed method has usefulness for removing the contaminated soil on the forest slope and for the reduction of the contaminated soil by separating the soil part in which cesium is mainly involved. On the other hand, the assumed contaminated soil in this test is different from actual complicated soil. Therefore, universal knowledge is not necessarily obtained only by this experiment. For example, the soil such as the decomposed granite soil has a complicated soil structure and contains a large amount of clay. The clay may have a frayed edge, and a lot of cesium may be adsorbed into the frayed edge. In near future, it is necessary to examine whether the decontamination and the volume reduction by the proposed method are possible for the soil such as the decomposed granite soil.

## 6. ACKNOWLEDGMENTS

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