

## STUDY ON REMOVAL METHOD OF CESIUM FROM SLOPE SOIL USING SIMPLE SLUSHER

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**ABSTRACT:** Though 6 years passed since the accident at the Fukushima Daiichi nuclear power station, and the decontamination of the difficult-to-return zone is not in prospect. The reason why the decontamination has not carried on is that of the geographic factor around the zone. In particular, there is a lot of forest area around the zone. Since the accident, radioactive cesium has been accumulated by constant wind and rainfall. Therefore, it is necessary to decontaminate the slope as it is the supply source of radioactive cesium from the forest to the residential area or farmland. Thus, in this study, the simple method of using running water was proposed to solve the decontamination of the forest area. This method proves that the vermiculite strongly fixes the radioactive cesium and the decontamination site is a slope. As a result, it is confirmed that the removal of vermiculites reduces 20% of the volume of contaminated soil. The vermiculite is easy to remove from the water since it is lighter in weight than sand and water. Removal of vermiculite using this method leads to volume reduction of the contaminated soil.

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

### 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 for all of the mentioned backgrounds. The radioactive materials were spread to the atmosphere by rain and breeze across mountains and rivers. The radioactive cesium was specifically detected in Tokyo Bay. Also, the radioactive materials that have spread across mountains are firmly bonded to the clay foods in the soil. Furthermore, the cesium-137 was extensively detected in Fukushima prefecture because it was vigorously discharged for some time. The adverse effect was strongly shown within the 1F, and the 12 municipalities in Fukushima prefecture was designated as the evacuation zone (see Fig. 1). Fig. 1 was developed based on the data by the Reconstruction Agency as in [1]. Okuma town, where the 1F is located, was declared the most difficult zone to restore among the municipalities. In Okuma town, the current air dose is lower than the air dose at the time, with the decreasing of cesium-134 (the half period is two years). However, the cesium-137 continuously emit the radioactivity

because the half period of cesium-137 is about thirty years. Therefore, it is considered to be difficult to decrease the radiation dose. The specific plans in the evacuation zone are not currently devised regardless of the unpredictable decrease in radiation dose. For example, the interim storage facilities are built in the Okuma to store the flexible container bag in which the contaminated soil is filled. The amount of contaminated soil estimated is 16~20 million cubic meters; and to secure interim storage facilities, where the contaminated soil is stored, is deemed difficult. Therefore, the volume reduction of the contaminated soil is an urgent business. Alternatively, the contaminated area is dominated by the mountains and village forests. Even if the radioactive cesium of flatland is removed, the clay particle including the radioactive cesium continues to be carried from the village forests by wind or rain as in [2]. Thus, the re-contamination is by the radioactive cesium is a concern. In this way, the slope of the mountainous area serves as a supply route of the radioactive cesium from the forest area to the residential areas and agricultural lands as in [3], [4].

Therefore, it is necessary to remove the radioactive cesium from the soil on the slope to reduce further collection contamination. The previous study is conducted by the faculty of engineering, Ibaraki University in order to settle this problem, and it has two features as in [5].

1. The bentonite is spread onto the leaf mold in the forest so as to control the resorption of the radioactive cesium to plants.

2. 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 [6]. 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. Thus, in this study, the simple decontamination method is proposed in order to decontaminate the contaminated soil on the slope by flowing water. The surface soil including a lot of cesium [7] is collected in this method, and it is suitable for the type of soil in Fukushima. The cesium is easily absorbed into the vermiculite which is eventually distributed in the soil of Fukushima. The specific weight of vermiculite is very small, and the vermiculite floats on water. Consequently, the vermiculite can be separated from the washed soil. Alternatively, both the decontamination and volume reduction of contaminated soil are conducted; because the cesium, which is not contained in the vermiculite, can be washed and collected. In this study, the reproducible slope test in which the Toyoura standard sand and vermiculite

are utilized to conduct and examine the usability of the proposed method.

## 2. CESIUM IN THE SOIL

A large number of radioactive materials was released by the accident in the 1F. Table 1 is the total amount of discharged radioactive materials in large quantities and its half-life. Table 1 is developed based on the date by Ministry of Economy as in [8]. The radioactive cesium has harmful long-term effects due to its long half-life. The volume of strontium-90 spreading 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

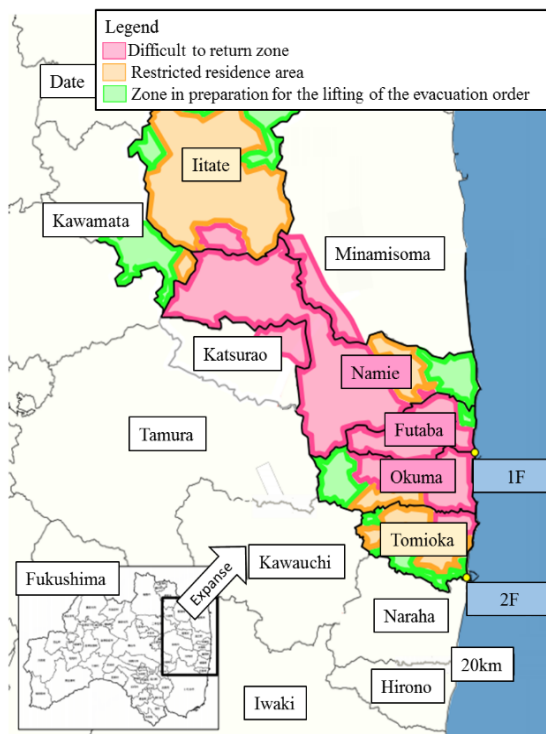


Fig. 1 Conceptual diagram in areas under evacuation orders as in [1]

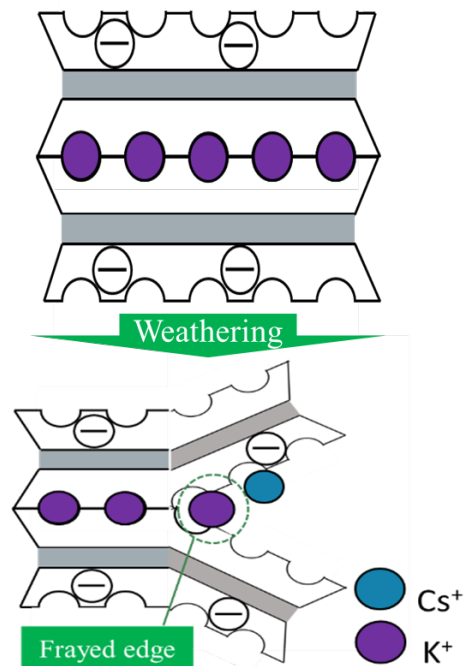


Fig. 2 Top part of 2:1 types of lamellar silicate and bottom part of Frayed edge

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. 2 of top 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. 2 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.

Table 1 Compendium of the discharged radioactive materials

Nuclide	Half-life (day)	Total discharge (Bq)
Xe-133	5.2	$1.1 \times 10^{19}$
Cs-134	766.5	$1.8 \times 10^{15}$
Cs-137	10950.0	$1.5 \times 10^{16}$
Sr-90	10621.5	$1.4 \times 10^{14}$
Ba-140	12.7	$3.2 \times 10^{15}$
Te-132	3.3	$8.8 \times 10^{16}$
I-131	8.0	$1.6 \times 10^{17}$
Sb-127	3.9	$6.4 \times 10^{15}$

### 3. MOCK CONTAMINATED SOIL

In this study, the Toyoura standard sand, vermiculite or their mixed sand is used as the mock contaminated soil that the cesium is added. The Toyoura standard sand consists of uniform size of soil grain (see Fig. 3). Moreover, it is very smooth, and the soil grains hardly stick to each other. Therefore, the difference between experimental samples can be neglected under the same condition, and the reproducibility of experiments are secured. The vermiculite is the silicate mineral that mainly consists of the oxidized silicon, magnesia and aluminum oxide. The vermiculite expands when heated at a high-temperature. 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 from a soil. In this study, the vermiculites of three-grain size are used so as to examine the influence of the different size of the grain on the adsorption of vermiculites. The actual soil consists of not only vermiculite but also the various minerals whose characterizations are different each other. The

actual soil environment is different from the artificial one when the mock contaminated soil consists of only the vermiculite. Therefore, the Toyoura standard sand and the vermiculite are mixed as the mock contaminated soil and the mixed soil is used in the experiments.



Fig. 3 Toyoura standard sand (upper left), vermiculite of 0.7mm (upper right), 1.0mm (lower left), 2.0mm (lower right)

### 4. TEST AND ANALYSIS METHOD

In order to imitate the real behavior of cesium in the soil slope, the slope device is produced (see Fig. 4). 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). 8.0 mm in diameter of the drain is set at the bottom of slope section in order to collect the water and soil which contains cesium. Also, the slope angle is set to about 9 degrees to avoid the flow disruption. The flowing water section is made by connecting the porous rubber hose to the water pump with a tube. The magnified drawing of the porous pipe is shown in Fig. 5. The porous pipe is made of the porous stone having many tiny pores from which the water flows away. The water flows in through the section (a) of the pipe and flows out from both the sections (b) and (c) of the pipe. It is needed to flush uniformly on the slope in this study. Thus, section (b) is blocked to keep the water from flowing out the section (b), which enables the water to flow out from only the section (c).

The experiment and analysis methods are explained below in detail. First, the cesium chloride is added to the assumed soil, and the assumed soil is set on the slope of the device. Secondly, the pure water is disembogued from the porous hose set on top of slope until the assumed soil is completely done away from the slope. Even if the pipe is processed as mentioned above, a few sample soils cannot be flushed and remain on the slope of the experimental device. The porous pipe is moved from right to left so as to flow the whole sample soil.

Finally, both pure water and soil are collected, and the cesium concentration is analyzed by an AAS (Atomic Absorption Spectrometer). The sample must be in a liquid state when the sample is analyzed by the AAS. Therefore, it is necessary to wash out the cesium in the soil after the test. As the washing out method, 200.0 mL of pure water is soured for the collected soil, and the sample is screwed with 300 rpm for the 10 minutes by the stirrer. After that, the solid-liquid separation is conducted four times with 38  $\mu\text{m}$  of sieve size. The extracted volume of cesium contained in both the solution and the soil after the test is quantitated for each filtrate by the AAS. Alternatively, in this study, all of the tests are conducted three times under the same conditions to ensure repeatability.

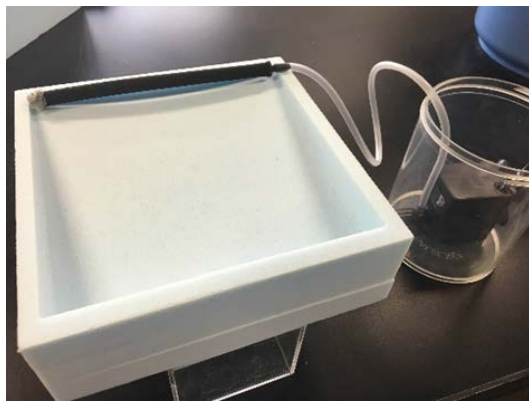


Fig. 4 Slope device for cesium behavior demonstration



Fig. 5 Magnification of porous pipe

## 5. PRELIMINARY TEST

The three preliminary tests are conducted at the previous step of slope test. One is the test in which only cesium is flushed out, another is the test in which only standard sand is flushed out, and the other is the test in which only vermiculite is flushed out. As discussed later, the methods and results of each test are explained in detail.

### 5.1 Cesium flush test

It is necessary to confirm whether added cesium remains or not on the slope after flushing and all cesium can be recovered or not in order to verify the accurate extraction amount of cesium. To begin with, 0.1 mL of cesium chloride solution is added to the slope and flushed out with pouring 100.0 mL of pure water over the slope. The amount of cesium in the recovered solution is analyzed by the AAS after the cesium is flushed out. The addition amount of cesium is 100.0  $\mu\text{g}$  (1 mg/mL). The extraction amount of cesium in the test is shown in Fig. 6. From the figure, an average 102.3  $\mu\text{g}$  of cesium is detected. It can be said that all cesium is flushed out from the addition amount of cesium. Consequently, it is confirmed that all cesium can be flushed out the slope in this method.

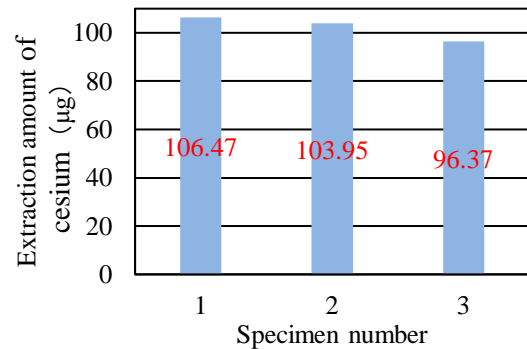


Fig. 6 Extraction amount of cesium in cesium flush test

### 5.2 Standard sand flush test

The assumed contaminated soil that mixes with the standard sand and the vermiculite is employed in the slope test discussed in the next section. In this subsection, the flush tests for the standard sand and the vermiculite are conducted in order to ascertain their adsorption properties of cesium. As for the standard sand, 5.0 g and 15.0 g of specimens are prepared. The pure water is poured over the slope until the standard sand in which 0.1 mL of cesium chloride solution is added is completely flushed out. The extraction amount of cesium in the solution is analyzed after collection and stirring by the stirrer.

First, the extraction amount of cesium from 5.0 g of standard sand is shown in Fig. 7. The extraction amount of cesium after flushing is an average 21.32  $\mu\text{g}$ , and about twenty percent of cesium is extracted against the addition amount. In addition, an average 35.6  $\mu\text{g}$  of cesium is detected from the solution after stirring, and more than fifty percent is extracted from 5.0 g of standard sand. Secondly, the extraction amount of cesium from 15.0 g of standard sand is shown in Fig. 8. An average 25.8  $\mu\text{g}$  of cesium is detected from the solution after

flushing. This result is more or less same as that of the test using 5.0 g of standard sand. On the other hand, an average 46.9  $\mu\text{g}$  of cesium is detected from the solution after stirring of the recovered sand and the extraction amount is higher than that of the test using 5.0 g of standard sand. It is confirmed that more than seventy percent of cesium can be extracted from the recovered solution and sand in the test using 15.0 g of standard sand, and the extraction amount in the test using 15.0 g of standard sand is slightly larger than that in the test using 5.0 g of standard sand. When an amount of sample is too small, the sample may accumulate on the bottom of the container and stirring may not be performed well.

Therefore, in the case of a small amount of sand, the wash away of recovered sand is not enough. In the future, additional provisions such as the increase of rotation speed of stirring are required.

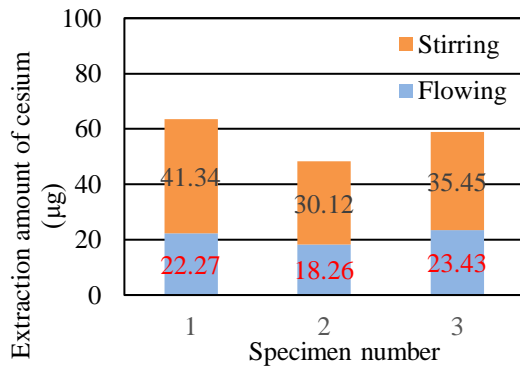


Fig. 7 Extraction amount of cesium in standard sand flush test (5.00g)

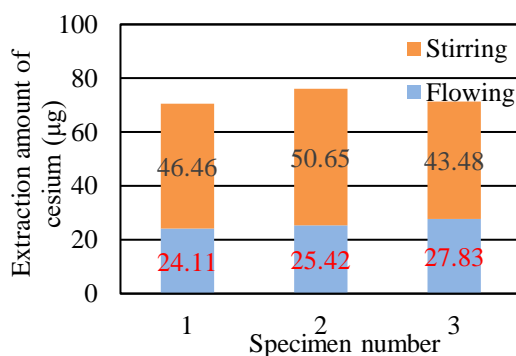


Fig. 8 Extraction amount of cesium in standard sand flush test (15.00g)

### 5.3 Vermiculite flush test

1.0 g of the vermiculite whose particle size is 0.7 mm, 1.0 mm or 2.0 mm is set on the slope and 0.1 mL of cesium chloride solution is added to the

vermiculite. Since it is conceivable that the vermiculite may consist of various particle sizes in the actual one, three particle sizes of the mixed vermiculite (hereinafter referred to as Mix) that three particle sizes of vermiculite are prepared. Besides the mass of each vermiculite is set to be more or less same in the test.

The extraction amount of cesium from each particle size of vermiculite is shown in Fig. 9. The extraction amount of cesium is an average 11.0  $\mu\text{g}$  from 0.7 mm of vermiculite, 13.2  $\mu\text{g}$  from 1.0 mm, 14.8  $\mu\text{g}$  from 2.0 mm and 13.2  $\mu\text{g}$  from the Mix. The extraction amount of cesium from the different particle size of vermiculite is more or less the same, though it increases in proportion as the particle size also increases. In addition, it is reconfirmed to be extremely difficult for the vermiculite to extract the cesium compared with the standard sand.

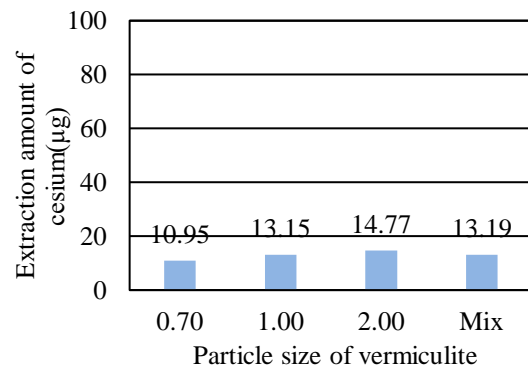


Fig. 9 Extraction amount of cesium in vermiculite flush test

## 6. ASSUMED CONTAMINATED SOIL FLUSH TEST

### 6.1 Test results

The test which an assumed contaminated soil is flushed out on the slope is conducted in order to make the assumption that the contaminated soil including the cesium on the slope is decontaminated. The assumed contaminated soil is prepared by mixing the standard sand with 1.0 g of the Mix. Besides, three specimens of 10.0 g, 15.0 g, and 30.0 g are prepared to regard to the standard sand, and each test result is compared.

First, the extraction amount of cesium from the assumed soil using 30.0 g of standard sand is shown in Fig. 10. No more than an average 15.3  $\mu\text{g}$  of cesium is extracted by flushing the soil. By contrast, the total extracted amount of cesium is 46.6  $\mu\text{g}$  on average which is added to the extracted amount from the solution after stirring. There is the same tendency between the test using the MIX and the test using only standard sand.

Secondly, the extraction amount of cesium from the assumed soil using 15.0 g of standard sand is shown in Fig. 11. An average 14.8  $\mu\text{g}$  of cesium is detected from the solution after flushing, and an average 32.1  $\mu\text{g}$  is detected from the solution after stirring the soil. The total extracted amount is 46.9  $\mu\text{g}$  on average, which is almost same as the extracted amount of the test using 30.0 g of standard sand.

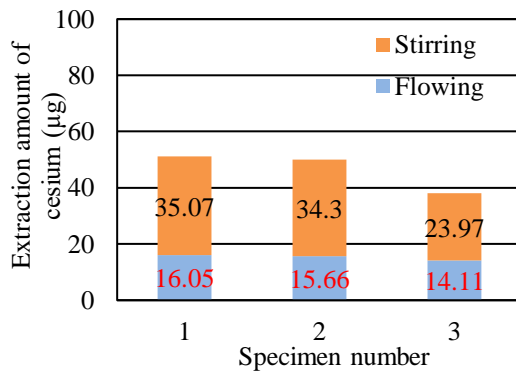


Fig. 10 Extraction amount of cesium from assumed soil using 30.0 g of standard sand

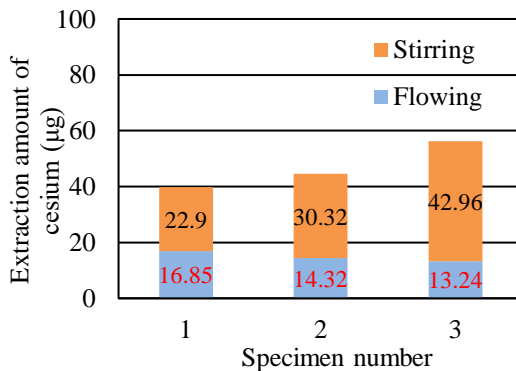


Fig. 11 Extraction amount of cesium from assumed soil using 15.0 g of standard sand

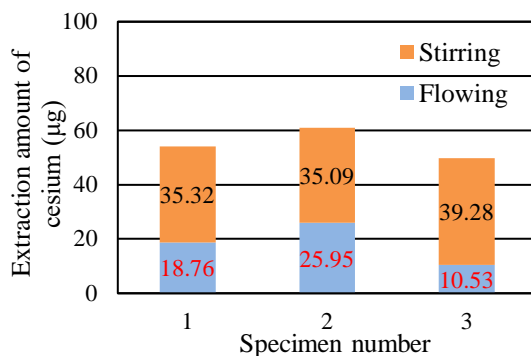


Fig. 12 Extraction amount of cesium from assumed soil using 10.0 g of standard sand

Finally, the extraction amount of cesium from the assumed soil using 10.0 g of standard sand is shown in Fig. 12. The extracted amount of cesium from the solution after flushing and stirring is 55.0  $\mu\text{g}$  on average. In addition, it is confirmed that about fifty percent of cesium is extracted from the assumed soil against 1.0 g of additive cesium regardless of the amount of standard sand. The cesium is extracted from the standard sand, but it is not extracted from the vermiculite. From these results, it is considered that the vermiculite including cesium can be recovered by flushing the contaminated soil out the slope with flowing water and the other cesium can be extracted by just flowing the water into the contaminated soil.

## 6.2 Separation and collection of soil after the test

The experimental results mentioned in the previous subsection indicates the potential of the volume reduction of the soil contaminated by a radioactive material when the volume reduction is realized by the collection of the vermiculite containing cesium and the standard sand through which cesium is extracted. Focusing on the vermiculate features, the vermiculate floats on the water since the density of vermiculite is smaller than that of water or sand. Thus, the vermiculite, sand, and water can be effectively separated and collected.



Fig. 13 Vermiculite floating on the water surface

## 7. CONCLUSIONS

In this study, the simple decontamination method is proposed in order to decontaminate the contaminated soil including cesium on the slope of forest or mountain area, and the flush test on the

slope is conducted to examine the performance and availability of the proposed method by which the cesium is removed from the slope. Specifically, the cesium is added into the assumed contaminated soil in which standard sand and vermiculite are mixed, and the assumed contaminated soil is flushed out from the slope by flowing water. As a result, the extraction ratio is about 25 % of extracted cesium to the additive amount of the cesium which is added to only the standard sand through the cesium is hardly extracted from only the vermiculite by the proposed method. Therefore, about 70 % of cesium from the standard sand and about 10% from the vermiculite can be extracted if each sample sand is stirred after the flush test. Alternatively, about 15 % of cesium is extracted by only flushing and about 50 % of cesium is done by stirring from soil that the standard sand and the vermiculite are mixed. While more amount of cesium can be extracted from sand soil by flushing and stirring, cesium is hardly extracted from clay mineral such as vermiculite by flushing as well as stirring. Thus, the clay mineral from which it is hard to extract cesium is directly collected and moved to the temporary storage facilities. On the other hand, it is encouraged to extract cesium from the sand soil by stirring before collecting. In this way, the cesium can be collected by selecting the efficient disposal methods for each sample sand. Furthermore, it is considered to lead to the validated volume reduction of contaminated soil.

## 8. FUTURE TASKS

### 8.1 Improvement of slope device

The generation of irregularity of the test time and the water dosage cannot be avoided when the sample sand is completely flushed in the existing slope device. The existing device has to be improved so as to prevent the sample sand from remaining and ensure the repeatability of tests. It is invented the improved plan that the flowing section is changed from the porous pipe to the polyvinyl chloride (PVC) pipe having the spiral holes. In concrete terms, the rotating shaft is set the top of the

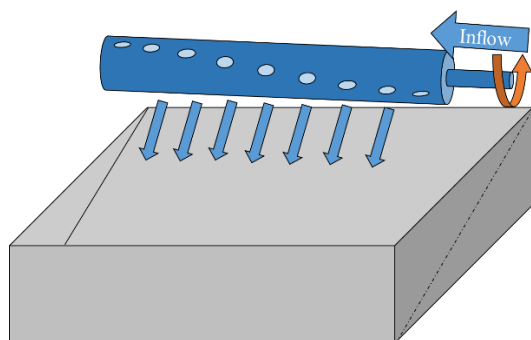


Fig. 14 Improvement plan of slope device

slope, and the PVC pipe and the rotating shaft are connected. After that, the PVC pipe is rolled at any rate. It is expected that the water is evenly flowed out from each hole by rolling the PVC pipe. (see Fig.14).

### 8.2 Improvement of simulated soil

The vermiculite and Toyoura standard sand are used as the soil that the composition is not complex in order to ensure the repeatability of the test in this study. Therefore, the separation of the sand and the vermiculite after the test is easy. On the other hand, the real soil has the complex composition. Thus, it is necessary to use the decomposed granite soil in the test. Furthermore, the separation and collection method may be examined in near future.

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