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Fundamental study on volume reduction of cesium contaminated soil by using magnetic force-assisted selection pipe

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Abstract

Advanced classification of Cs contaminated soil by using a magnetic force-assisted selection pipe was investigated. A selection pipe is a device that sort particles depending on their particle size, based on the relationship between buoyancy, drag, and gravity force acting on the particles. Radioactive cesium is concentrated in small-particle size soil components with a large specific surface area. Hence, the volume of the Cs contaminated soil can be reduced by recycling the large-particle size soil components with low radioactive concentration. One of the problems of the selection pipe was that the radioactive concentration of the stayed soil in the selection pipe exceeds 8000 Bq/kg, which is the standard value of recycling of Cs contaminated soil, due to low classification accuracy. In this study, magnetic fields were applied to the lab-scale selection pipe from upper side to improve the classification accuracy and to reduce the radioactive concentration of the stayed soil.

Keywords: radioactive cesium, advanced classification, selection pipe, volume reduction, halbach magnet

1. INTRODUCTION

In March 2011, the Great East Japan Earthquake caused the Fukushima Dai-ichi Nuclear Power Plant accident. Approximately 14 million m^3 of removed soil (hereinafter referred as "Cs contaminated soil") was discharged as a result of the accident [1]. Since the storage and management of the large amount of Cs contaminated soil need a lot of space and cost, the volume reduction techniques of Cs contaminated soil have been needed.

Fig. 1 shows the flow chart of the volume reduction of the Basically, normal wet or dry classification at 75 µm can sort the contaminated soil into high and low concentration components. However, agricultural soil mainly consists of clay and silt components under 75 µm, and only normal classification cannot reduce the volume sufficiently. Here, we propose an advanced classification process using selection pipes assisted by magnetic force for small particle size components under 75 µm in Soil B (the soil with 8000 to 20000 Bq/kg in radioactivity concentration), among the classification by the Ministry of the Environment in Japan[1]; "Soil A" is under 8000 Bq/kg, "Soil B" is 8000 to 20000 Bq/kg, "Soil C" is 20,000 to 80,000 Bq/kg, and "Soil D" is over 80,000 Bq/kg. The reason why we deal with Soil B is that the radioactive concentration is relatively low among Cs contaminated soil, and the

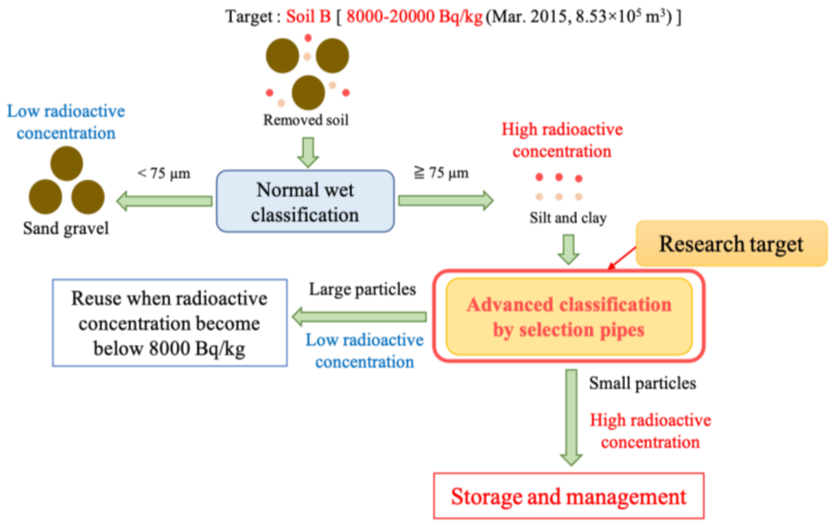

Fig. 1. Volume reduction flow of Cs contaminated soil and the target process in this study.

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classification point (the boundary particle size in the classification) can be set lower in the advanced classification process, which will result in high-volume reduction rate. Our aim was set to reduce total 50% of the volume of the soil components under 75 µm (silt and clay), obtained by normal wet classification.

In the advanced classification by the selection pipe, the soil suspension flows from the bottom to the top of the tube at a constant rate, and the components stayed in the pipe (large particle components) will be recycled only when the radioactive concentration become below 8000 Bq/kg. In our previous study, normal advanced classification by the selection pipe was conducted for small particle size components under 75 µm after normal wet classification, but the radioactive concentration of the stayed soil in the selection pipe exceeded 8000 Bq/kg (the standard value of recycling of Cs contaminated soil), due to low classification accuracy. There is a possibility that small clay particles aggregate each other or adhere to large particle, but the classification accuracy itself was low even in sufficiently dispersed model soil, because large particles will inhibit the ejection of small size particles. Hence, in this study, we investigated a method to reduce the radioactive concentration of the soil stayed in the selection pipe further, by assisting the soil separation by magnetic force applied from upper side of the pipe.

2. BASIC CONCEPT OF ADVANCED CLASSIFICATION BY A SELECTION PIPE

Fig. 2 shows a schematic diagram of the selection pipe system and the forces acting on the particle.

A selection pipe is a device that can sort the particles depending on their size, based on the relationship between buoyancy, drag, and gravity force acting on the particles flowing in the selection pipe [2]. The particles flowing in the selection pipe, from the bottom to the top, are subjected to buoyancy, drag, and gravity force. The resultant force of these three forces acting on the particles is shown in the equation (1) (the downward vertical direction is positive).

$$\mathbf{F} = \frac{4}{3}\pi r^3(\rho_p - \rho_f)\mathbf{g} - 6\pi\eta r(\mathbf{V_f} - \mathbf{V_p}) \quad (1)$$

Here, r is the particle radius [m], ρ_p is the density of the particle [kg/m^3], ρ_f is the density of the fluid [kg/m^3], $\mathbf{g}$ is the acceleration of gravity [m/s^2], η is the viscosity of the fluid [Pa・s], $\mathbf{V_f}$ is the velocity of fluid [m/s], and $\mathbf{V_p}$ is the velocity of the particle [m/s]. In equation (1), the velocity of the particle when the velocity of fluid ($\mathbf{V_f}$) is equal to zero is called the terminal velocity ($\mathbf{V_s}$) shown in equation (2).

$$\mathbf{V_s} = \frac{2r^2(\rho_p - \rho_f)\mathbf{g}}{9\eta} \quad (2)$$

When the terminal velocity is larger than or equal to the

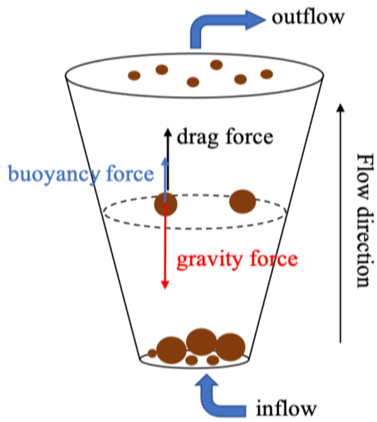

Fig. 2. A schematic diagram of the selection pipe and the forces acting on the particle. (An example of a conical selection pipe with different velocity distributions.)

When the terminal velocity is larger than or equal to the fluid velocity ($\mathbf{V_s} \geq \mathbf{V_f}$), the particles settle in the selection pipe against the fluid flow and stay in the selection pipe. On the other hand, when the terminal velocity is less than the fluid velocity ($\mathbf{V_s} < \mathbf{V_f}$), the particles are ejected from the upper side of the selection pipe by the fluid flow. The classification point can be controlled by inflowing the fluid into the selection pipe so that the fluid velocity $\mathbf{V_f}$ in the pipe is equal to the terminal velocity of the particles $\mathbf{V_s}$, which corresponds to the classification point. As a result, the large particles above the classification point stay in the selection pipe, whereas the small particles below the classification point are ejected from the selection pipe.

Since most of the radioactive cesium is adsorbed on the surface of the soil particles, the amount of radioactive cesium adsorbed per unit mass highly depends on the particle size of the soil, corresponding to the specific surface area. Based on this, the soil stayed in the selection pipe is low radioactive concentration due to its large particle size, whereas that ejected from upper side is high radioactive concentration due to its small particle size. Finally, the volume of Cs contaminated soil can be reduced by recycling the stayed soil.

3. METHOD TO IMPROVE SEPARATION ACCURACY BY USING A HALBACH MAGNET

Here, we describe a method to improve the separation accuracy in order to reduce the radioactive concentration of the stayed soil by using magnetic force.

Clay minerals are generally classified into 1:1 and 2:1 type of clay minerals. In general, it is known that 1:1 type clay minerals are diamagnetic and 2:1 type clay minerals are paramagnetic. Table 1 shows the classification of major clay minerals in the soil and its volume magnetic susceptibility. It is also known that 2:1 clay minerals strongly immobilize considerably more cesium than 1:1 clay minerals, due to the difference in their layer structure [3].

The magnetic force ($\mathbf{F_M}$) acting on a particle placed in a magnetic field is expressed as equation (3).

TABLE 1
CLASSIFICATION OF CLAY MINERALS AND ITS VOLUME MAGNETIC SUSCEPTIBILITY.

Types of minerals		Volume magnetic susceptibility (-)
1:1 type clay	Kaolinite	-2.0×10^{-5}
2:1 type clay	Vermiculite	2.6×10^{-4}
	Illite	3.1×10^{-4}
	Montmorillonite	8.2×10^{-5}

$$\mathbf{F_M} = V(\mathbf{M}\cdot\nabla)\mathbf{B} \tag{3}$$

Here, V is the volume of the particle [m^3], $\mathbf{M}$ is the magnetization of the particle [T], and $\mathbf{B}$ is the magnetic flux density at the position of the particle [T]. It can be seen in equation (3) that the magnetic force acts in the direction of attraction to the magnet for 2:1 type of clay minerals because of their paramagnetism. On the other hand, magnetic force acts in the direction away from the magnet for 1:1 type of clay minerals because of their diamagnetism.

Based on this, a magnet was placed near the outlet at the top of the selection pipe, so that magnetic force was exerted on the soil particles in the selection pipe and promote the ejection of 2:1 type of clay minerals from the pipe, thereby reducing the radioactive concentration of stayed soil.

4. EXPERIMENTAL METHOD

Fig. 3 shows the lab-scale selection pipe used in this study. In this experiment, vermiculite with particle sizes of less than 75 μm were used as a model soil, assuming the fine grain content (clay and silt) after normal wet classification. As shown in Table 1, vermiculite is classified as a 2:1 type of clay mineral. As described in the previous section, 2:1 type of clay minerals are paramagnetic and adsorb more radioactive cesium than 1:1 clay minerals and silt (mainly quartz and feldspar). If the ejection rate of highly radioactive 2:1 type of clay minerals from the selection pipe is promoted by using the magnet, it can be expected to increase the effectiveness of reducing the radioactive concentration of the stayed soil.

Hence, we conducted advanced classification experiments of vermiculite particles by using a Halbach magnet in order to investigate the effectiveness of applying a magnetic field from on 2:1 type of clay minerals. Here, we did not use a mixed system with kaolinite, a typical 1:1 clay mineral, because it is difficult to measure the particle size distribution of each component before and after advanced classification in a mixed system.

A Halbach magnet is assembly of several permanent magnets arranged so that the magnetic field is concentrated on one side of the magnet to obtain a large magnetic flux density [4]. Here we used a Halbach magnet (231 mm in length, 128 mm in width, and 60 mm in height, manufactured by NEOMAX Engineering, Co., Ltd.) which has a maximum surface magnetic flux density of about 1.3 T, and installed it directly above the selection pipe for advanced classification. The experimental system and magnetic field distribution around Halbach magnet are respectively shown in Fig. 4 and 5, and the experimental conditions are shown in Table 2. When the magnetic field is applied from the top of the selection pipe, the magnetic force will assist the ejection of paramagnetic 2:1 type clay minerals from outflow.

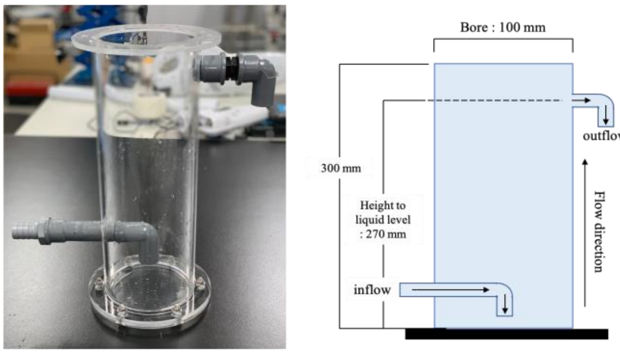

Fig. 3. The selection pipe used in this study.

The suspension of vermiculite particles with a solid-liquid ratio of 1:100 was prepared with distilled water, and set the classification point at 30 μm. Here, 30 μm of classification point and corresponding flow rate was determined based on the estimation of radioactivity concentrations of the actual contaminated soil after normal and advanced classification. We conducted the same experiment twice to confirm reproducibility.

Here, 30 μm of classification point and corresponding flow rate was determined based on the estimation of radioactivity concentrations of the actual contaminated soil after normal and advanced classification. Table 3 shows the initial radioactivity concentrations of the Cs contaminated soil and the estimated radioactivity concentrations of the classification points and stayed soil in the selection pipe, assuming that the classification accuracy is 100 % and that radioactive cesium is uniformly adsorbed and is simply proportional to the surface area of the soil particle, calculated from the measured particle size distribution. At the maximum concentration of 20,000 Bq/kg in soil B, advanced classification is effective, i.e. the soil after classification becomes under 8000 Bq/kg, when the classification point is set at 30 µm or more. The classification point was set at 30 µm based on this estimation.

A Halbach magnet was placed directly above the selection pipe putting the high-magnetic field side down, and then water was circulated through the tube at a flow rate of 0.30 L/min by a fixed volume pump. Then, 2.12 L of vermiculite suspension flowed into the selection pipe at a flow rate of 0.30 L/min. Here, the initial total volume of vermiculite suspension was set to the maximum capacity of the selection pipe (2.12L).

After 15 L of water was flown into the selection pipe, there was no temporal visible change in the turbidity of the suspension in the pipe, showing that the ejection of soil particles from the pipe was considered to be completed. As As soon as all the suspension was flowed, the distilled water was flowed into the selection pipe at the same flow rate (0.30 L/min), and when total 15 L of suspension was ejected, the pump was stopped to collect the suspension stayed in the selection pipe. a control experiment, the same

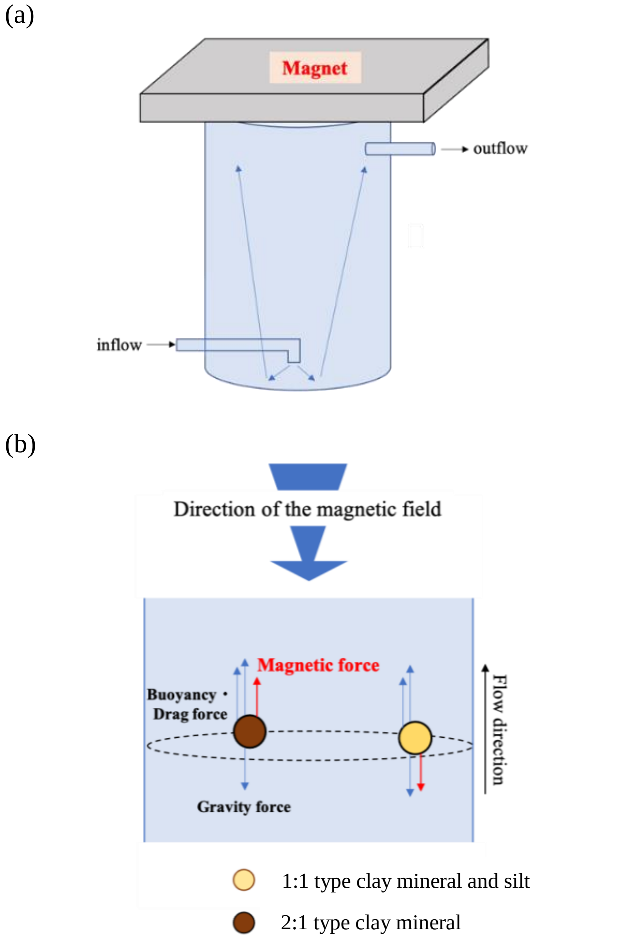

Fig. 4. Experimental system of magnetic force-assisted selection pipe; (a) setup of the advanced classification system, (b) the forces acting on the particles under the magnetic field.

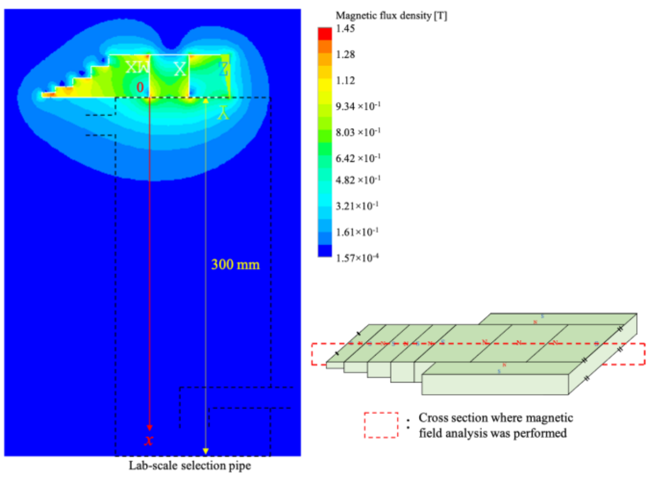

Fig. 5. The magnetic field distribution around a Halbach magnet calculated by finite element method.

TABLE 2
EXPERIMENTAL CONDITIONS OF THESE EXPERIMENTS.

Amount of vermiculite	Solid-liquid ratio	Classification point	Flow rate
21.2 g	1:100	30 µm	0.30 L/min

TABLE 3
INITIAL RADIOACTIVE CONCENTRATION OF THE SOIL AND ESTIMATED RADIOACTIVE CONCENTRATION OF STAYED SOIL AT EACH CLASSIFICATION POINT IN ADVANCED CLASSIFICATION.

Initial *1 [Bq/kg]	After normal classification [Bq/kg] *2	Advanced classification point [µm] *3				
		10	15	20	25	30
20000	59410	25503	16859	10787	9243	6666
19000	56439	24228	16017	10247	8781	6333
18000	53469	22953	15174	9708	8319	5999
17000	50498	21678	14331	9169	7857	5666
16000	47528	20403	13488	8629	7395	5333
15000	44557	19127	12645	8090	6932	4999
14000	41587	17852	11802	7551	6470	4666
13000	38616	16577	10959	7011	6008	4333
12000	35646	15302	10116	6472	5546	4000
11000	32675	14027	9273	5933	5084	3666
10000	29705	12752	8430	5393	4622	3333
9000	26734	11476	7587	4854	4159	3000
8000	23764	10201	6744	4315	3697	2666

*1; Soil B before classification
*2; Soil fractions smaller than normal classification point (75 µm)
*3; Stayed soil fractions larger than each advanced classification point

classification experiments under the conditions shown in Table 2 was conducted 3 times without a magnetic field. After the advanced classification, a small amount of the suspension stayed in the selection pipe and the suspension ejected from the selection pipe were respectively sampled, and the particle size distribution was measured using a laser diffraction/scattering particle size analyzer (LA-920, HORIBA, Co., Ltd.). We also measured the dry weights of each stayed and ejected particles.

5. RESULTS AND DISCUSSIONS

As described in the previous sections, the accuracy of classification and the radioactive concentration of stayed soil are important because the stayed soil is to be recycled after the advanced classification by the selection pipe. Thus, we focus on stayed soil in the results and discussions.

Fig. 6 shows the comparison of the classification accuracy of the stayed soil between the experiments with and without the magnet. The vertical axis shows the classification accuracy of the stayed soil, which is defined as the volume fraction of particles above the classification point (30 µm) in the total stayed soil, calculated from the results of particle size distribution. Fig. 7 shows the particle size distribution of stayed/ejected soils with and without magnet. The results in Fig. 6 was calculated by the integral of volume fraction above 30 µm.

It was shown from Fig. 6 that application of magnetic field with a Halbach magnet improved the classification accuracy of the stayed soil by 5.8 %. The results indicate that application of magnetic force on the vermiculite particles is effective in promoting the ejection of vermiculite particles below the classification point of 30 µm from the selection pipe.

Based on these results, we estimated the radioactive concentration of the stayed soil, in order to confirm the effectiveness of reducing the radioactivity concentration of the stayed soil by the magnetic force. In this estimation

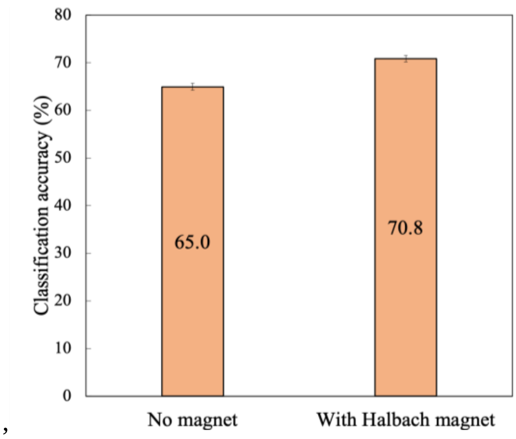

Fig. 6. Classification accuracy of stayed soil with and without Halbach magnet.

assuming that the radioactive cesium is uniformly adsorbed on the soil surface and the soil particle is the spherical shape, the radioactivity of the adsorbed radioactive cesium was calculated from the ratio of the surface area based on the particle distribution shown in Fig.7.

It was also assumed that the Cs contaminated soil includes 70 % of 2:1 type of clay minerals and 30 % of 1:1 type of clay minerals, based on the composition of clay minerals in the typical lowland soil in Fukushima Prefecture [5], and that 2:1 type of clay minerals adsorbs 1.13 times more radioactive Cs than 1:1 type of clay minerals. In the calculation, we assumed that the specific surface area of 2:1 type of clay minerals was 1.13 times larger than that of 1:1 type of clay minerals of the same particle size [6]. The 1:1 type clay minerals were assumed to have the same particle size distribution as the vermiculite used in this experiment, and was assumed to be subject to a magnetic force in the direction away from the magnet depending on their particle size. The radioactivity concentration of the soil after advanced classification was calculated by dividing the radioactivity by the soil dry weight.

The estimation results are shown in Fig. 8. It was shown that the radioactive concentration of the stayed soil could be reduced by 20 % compared to the case without the magnet.

This indicates that the application of a magnetic field can assist the ejection of paramagnetic 2:1 type of clay minerals from the selection pipe, which adsorb a large amount of radioactive cesium, can enhance the reduction of the radioactive concentration of the stayed soil.

The reason why the effect of the Halbach magnet is greater in the composite system of 1:1 and 2:1 type than in single system of 2:1 type is thought to be that the magnetic field acts in the opposite direction for paramagnetic 2:1 type and diamagnetic 1:1 type clay minerals. The high-dose component, vermiculite, is gravitated upward and ejected by the magnet, while the low-dose component, kaolinite, is weakly dragged downward by the magnet. This leads to a greater increase in the radioactivity concentration of the ejected fraction, and thus the radioactive concentration of the stayed fraction relatively decreases by the magnet. As a result, the soil component with high radioactive concentration is selectively ejected

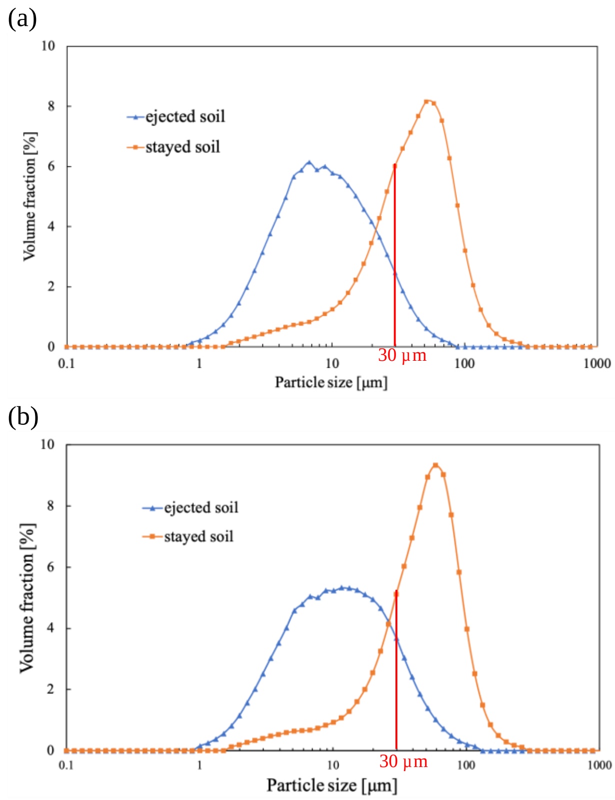

Fig. 7. Particle size distribution of ejected/stayed vermiculite; (a) without and (b) with Halbach magnet.

from the selection pipe, which makes the classification accuracy high.

In this study, a Halbach magnet was used to apply a strong magnetic field from the top of the selection pipe easily, but the objective value less than 8000 Bq/kg will not be achieved as shown in the estimation in Fig. 8 because of the range of magnetic field was small. Effective improvement in classification accuracy can be expected by applying a magnetic field from upper side of the selection pipe with a superconducting bulk magnet or by inserting the top of the selection pipe into the bore of superconducting solenoidal magnet. In the future, we plan to study the practical advanced classification system using actual contaminated soil and a superconducting magnet.

We have also studied the volume reduction of Cs contaminated soil using a high gradient magnetic separation (HGMS) system with superconducting solenoidal magnets and magnetic mesh filters [7-10], but there were problems of slow processing speed and early saturation in the soil capture amount of the magnetic filters. The method in this study could be a new method to solve these problems of HGMS.

6. CONCLUSION

Advanced classification experiment with a selection pipe was conducted by using the Halbach magnet in order to investigate the effectiveness of applying a magnetic field to assist the ejection of 2:1 type of clay minerals from the selection pipe.

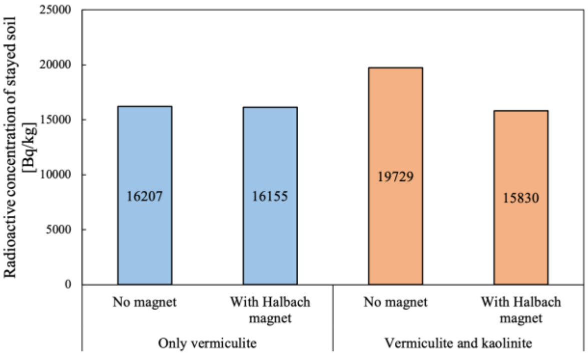

Fig. 8. Changes in the radioactive concentration of stayed soil by using the Halbach magnet.

The experimental results showed that the classification accuracy of the stayed soil was improved by 5.8 % for vermiculite particles.

The estimation of radioactive concentration based on the result indicated that selective ejection of paramagnetic 2:1 type of clay minerals from the selection pipe by magnetic force will enhance about 20% reduction of the radioactive concentration of the stayed soil.

In the future, we will conduct advanced classification experiments for actual Cs contaminated soil using bulk or solenoidal superconducting magnets in a scaled-up selection pipe system, in order to verify the effectiveness of reducing the radioactive concentration of the actual soil.

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