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Fundamental study on volume reduction of cesium contaminated soil using cyclone-type magnetic separator

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Abstract

A large amount of cesium-contaminated soil was generated as a result of the decontamination work following the accident at the Fukushima Daiichi Nuclear Power Plant. To reduce the final disposal volume of contaminated soil, it is necessary to separate the contaminated soil into low- and high-dose soil components and reuse the low-dose soil under 8000 Bq/kg. We have investigated a magnetic separation technique to reduce the volume of the contaminated soil. Magnetic separation is a volume reduction technology that utilizes these differences in magnetic properties. However, the high-gradient magnetic separation technique (HGMS) we have been studied has problems such as clogging of filters and low separation accuracy due to the passage of 2:1 type clay minerals with small particle diameters.

In this study, we propose a new separation method using a cyclone-type magnetic separator that focuses not only on magnetic susceptibility but also on differences in particle size. The cyclone-type magnetic separator can separate 2:1 type clay minerals from 1:1 type clay minerals by inducing 1:1 type clay minerals with large particle diameters to the outside of the cylinder and 2:1 type clay minerals with small and large particle diameters to the inside of the cylinder through the difference in the combined magnetic and centrifugal forces acting on soil particles. Separation accuracy was evaluated using simulated soil consisting of vermiculite and kaolinite. Based on these results, the reduction rate of the radioactivity concentration was estimated, and the design guidelines of the device for practical use were discussed.

Keywords: Fukushima Daiichi Nuclear Power Plant accident, cesium contaminated soil, volume reduction, cyclone-type magnetic separation, magnetic susceptibility, radiation concentration

1. INTRODUCTION

A large amount of soil contaminated with radioactive cesium was generated as a result of the decontamination work following the accident at the Fukushima Daiichi Nuclear Power Plant. The amount of soil removed as a result of decontamination work in Fukushima Prefecture is amounted to approximately 13 million m^3[1]. The volume reduction of this large amount of removed soil is being considered to reduce the final disposal volume and to reuse the low-dose soil. The main technologies considered by the Japanese government for the volume reduction of removed soil are classification, chemical treatment, and thermal treatment. According to the Japanese Ministry of the Environment standards, soil with a radioactivity level of 8,000 Bq/kg or less can be reused. Based on the Japanese Ministry of the Environment standard, soil with radioactivity below 8,000 Bq/kg is recyclable. This criterion was established based on a scenario assessment of the effects of radioactive materials to ensure that the doses received by residents and workers in the vicinity do not exceed 1 mSv/year, which is the effective dose limit for the general public [1].

Classification treatment technology is a method for separating soil into silt and clay (75 μm or smaller) and sand and gravel (75 μm or larger), based on the fact that radioactive cesium tends to adhere to silt and clay, which are small-diameter particles in soil. It has been used as a conventional technology to separate heavy metals with similar characteristics. Although it can treat large volumes of soil at relatively low cost, it has the problem that it cannot effectively reduce the volume of agricultural soil, which contains a large amount of clay and silt with small particle sizes [2].

Chemical treatment technology is a method of separation in which radioactive cesium in soil is eluted into the solvent using a strong acid, and cesium in the solution is collected by the adsorbents. The separation efficiency is high, and it is expected to be effective not only for sandy soil but also for clayey soil; however, it is expensive and requires the treatment of secondary wastes such as solvents and adsorbents [2].

Thermal treatment is a method of vaporizing radioactive cesium by adding a reaction accelerator to the soil and heating it to 1350℃ or higher. The vaporized radioactive cesium is cooled until it solidifies and is then collected using bag filters. It can be applied to both sandy and clayey soils and has a high separation efficiency, but it is expensive, requires a considerable amount of reaction accelerant depending on the material to be treated, and has issues in handling products with altered soil properties [2].

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Currently, the classification treatment technology has the highest level of technological maturity among these technologies. It can be used as a basic technology to treat large volumes of removed soil at low cost, and we are working to put it into practical use. The basic treatment process to reduce the volume of removed soil is to first remove foreign matters, and then to consider the use of volume reduction treatment alone or in combination [2]. The basic policy is to apply advanced treatments, such as thermal treatment, to fine-grained soil with relatively high radioactivity after classification.

Based on these issues of existing methods, we have proposed volume reduction of removed soil using magnetic separation technology as a new advanced treatment method. Magnetic separation technology can be applied to both sandy and clayey soils. Because it is a physical separation method that leaves cesium adsorbed in the soil, there is no need for strong acids or reaction accelerators, and the safety of the soil after separation is an advantage. A common method of magnetic separation is the high-gradient magnetic separation method (HGMS), in which a ferromagnetic magnetic filter is placed in a magnetic field and the object is captured on the filter. However, this method has problems such as clogging, the need for periodic cleaning, and the need for a strong magnetic field, making it difficult to ensure a practical processing volume and processing speed [3, 4].

In this study, we propose a new separation method using a cyclone-type magnetic separator as a new volume reduction method that uses magnetic force. This method can selectively separate 2:1 type clay minerals that are paramagnetic and adsorb large amounts of radioactive cesium from cesium-contaminated soil owing to the differences in the magnetic susceptibility of the soil components. The magnetic separation method has advantages over the three existing methods mentioned above, as it is a room-temperature separation method that uses physical remote forces. This method can be applied to particles with smaller diameters, in contrast to the classification method, does not require reagents such as strong acids or bases for chemical treatment, and does not require large amounts of energy such as heat treatment. Here, a laboratory-scale cyclone-type magnetic separation apparatus was fabricated as a basic experiment for such applications, and the feasibility of separating the simulated soil was investigated.

2. CONSEPT OF CYCLONE-TYPE MAGNETIC SEPARATION TECHNOLOGY

2.1. Magnetic Separation of Clay Minerals

Magnetic separation is a technology for selectively separating target substances from various mixtures and media by applying a magnetic force. A schematic view of a cyclone-type magnetic separation system was used for fine particles (silt and clay) of 75 μm or less after classification. Clay minerals contained in fine particles are classified into two types: 1:1 type clay minerals such as kaolinite and 2:1 type clay minerals such as vermiculite. The properties of each are listed in Table I [5].

TABLE I
CHARACTERISTICS OF CLAY MINERALS IN THE SOIL [5].

	1:1 type clay mineral	2:1 type clay mineral
volume magnetic susceptibility (-)	-2.0×10^{-5}	3.0×10^{-4}
Adsorption capacity of Cs (mmol/kg)	6	25,900

These two types of clay minerals have different cesium adsorption characteristics and magnetic properties, which can be used for magnetic separation. The 1:1 type clay minerals adsorb less Cs and are diamagnetic. In contrast, 2:1 type clay minerals adsorb more cesium and are paramagnetic. By applying a magnetic force to the removed soil, 2:1 type clay minerals could be selectively separated.

We have studied the use of high-gradient magnetic separation (HGMS). This technique traps paramagnetic soil particles on a ferromagnetic mesh filter that is placed in a magnetic field. Although the possibility of separation was demonstrated in principle, it was difficult to achieve a practical processing volume and processing speed because of issues such as filter clogging, periodic cleaning, and the need for a strong magnetic field and magnetic gradient. In this study, we propose a cyclone-type magnetic separation method that combines magnetic force and swirling flow.

The geometry of the magnetic separator is shown on the left side of Fig. 1. The soil suspension flowed in a circumferential direction from the top of a cylinder with an outer diameter of 77 mm and an inner diameter of 34 mm, generating a swirling flow inside the cylinder. This swirling flow exerts a centrifugal force on the soil particles in the outward direction of the cylinder. In addition, a magnet bar with multiple neodymium magnets was installed at the center of the cylinder to exert a magnetic force on the soil particles. The magnetic forces act in the outward direction of the cylinder for the diamagnetic 1:1 type clay minerals and in the inward direction of the

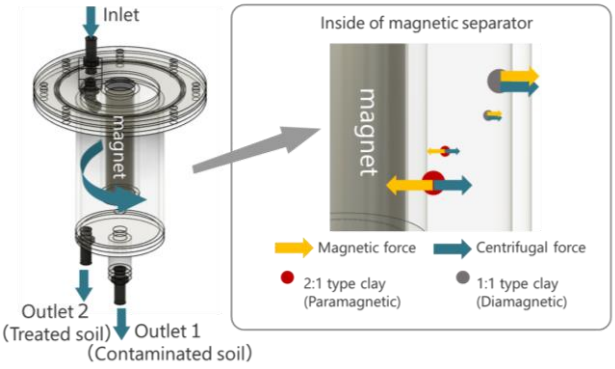

Fig. 1. A schematic view of cyclone-type magnetic separation system. The figure on the left is an overview of the apparatus, and the figure on the right shows the forces acting on the particles of 1:1 and 2:1 type clay minerals, respectively.

TABLE II
CALCULATION CONDITIONS OF MAGNETIC FORCE AND CENTRIFUGAL FORCE.

r	:	Particle radius (m)
r_A	:	Distance from center (m)
χ	:	Magnetic susceptibility of target particles (-)
μ_0	:	vacuum permeability
B	:	Applied magnetic field (T)
x	:	Wire Radius (m)
m	:	Particle weight (kg)
v	:	particle velocity (m/s)

cylinder for the paramagnetic 2:1 type clay minerals. These centrifugal and magnetic forces guide the larger 1:1 type clay minerals to the outside of the cylinder and the smaller and larger 2:1 type clay minerals to the inside of the cylinder. By providing two outlets, one at the center of the bottom and the other outside, a soil suspension with higher and lower radioactivity can be separated.

2.2. Estimation of Forces Acting on Soil Particles

The main forces acting on the soil particles in a cyclone-type magnetic separator are magnetic and centrifugal forces, as shown on the right side of Fig. 1. Buoyancy and gravity are neglected, assuming that they are balanced. The magnetic force F_M and centrifugal force F_C are represented by (1) and (2), respectively. Table II lists the numerical values of the equations. If the sum of the magnetic and centrifugal forces is positive, the particles are guided toward the center and discharged from Outlet 1. If it is negative, they are guided toward the wall of the cylindrical channel and discharged from Outlet 2.

$$F_M = \frac{4}{3}\pi r^3 \frac{\chi}{\mu_0}\frac{dB}{dx} \quad (1)$$

$$F_C = -mv^2 / r_A \quad (2)$$

Fig. 2 shows the results of calculating the magnetic and centrifugal forces acting on vermiculite and kaolinite based on (1) and (2). Here, the force acting on the magnet was assumed to be positive. The contribution of the magnetic force was large for vermiculite, and the contribution of the centrifugal force was slightly larger for kaolinite, although the magnetic and centrifugal forces were almost equal in order.

Fig. 3 shows the results of the calculation of the combined magnetic and centrifugal forces acting on vermiculite and kaolinite. Because the radial direction toward the magnet (toward the center) is positive, if the combined force is positive, the material is discharged from Outlet 1 at the center; if it is negative, it is discharged from Outlet 2 on the side. The calculation results showed that the combined magnetic and centrifugal forces were positive for vermiculite ejected from Outlet 1, and negative for kaolinite ejected from Outlet 2. Based on these results,

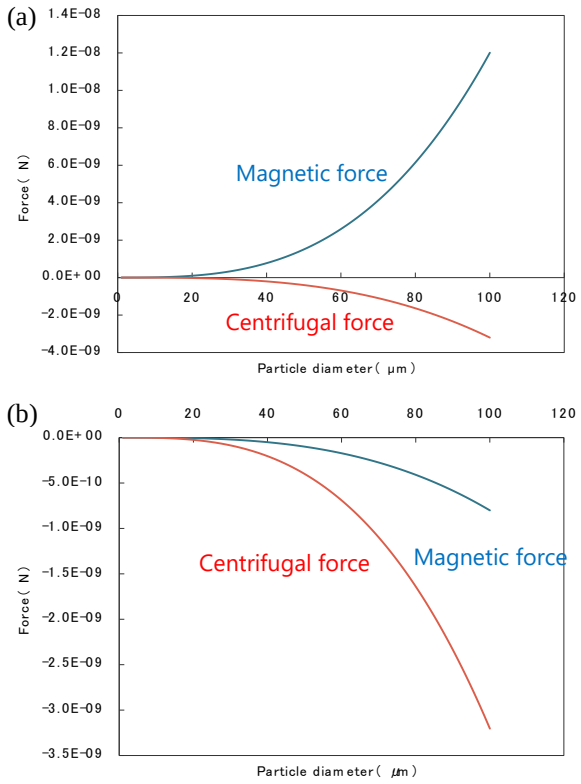

Fig. 2. Magnetic and centrifugal forces acting on the (a) vermiculite and (b) kaolinite particles.

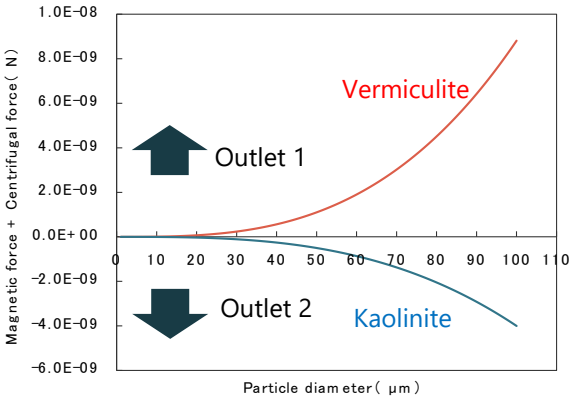

Fig. 3. Combined magnetic and centrifugal forces acting on vermiculite and kaolinite.

we predicted that the separation of 2:1 type clay minerals from 1:1 type clay minerals is feasible using a cyclone magnetic separation system. Based on these results, a laboratory-scale cyclone-type magnetic separation device was fabricated, and the feasibility of the separation was verified.

3. EXPERIMENTAL METHOD

3.1. Magnetic Separation Experiment

Vermiculite (SRVM FINE, Tomoe Kogyo Co., Ltd.) was crushed using a crusher (Wonder Crusher WC-3, Osaka Chemical Co., Ltd.) and dry-classified using a 75 μm sieve. A model soil suspension was prepared by

weighing 0.2 g of vermiculite and 0.3 g of kaolinite (ASP-400P, average particle size 3.5 µm, Tosin Kasei Co., Ltd.) and mixing it with 500 ml of distilled water in a plastic beaker. This is because the composition of clay minerals in Fukushima Prefecture is approximately 40% 2:1 type clay minerals and 60% 1:1 type clay minerals [6,7].

A schematic of the experimental procedure is shown in Fig. 4. For pretreatment, the soil suspension was sonicated using a tabletop ultrasonic cleaner (CPX1800-J, Branson) for 10 min and then stirred for 5 min at a speed of 6 (26,000 rpm) in a homogenizer (DIAX 900, Heidolph). This sonication and homogenization process is a rather intense agitation because the particles need to be well dispersed in water to ensure the separation of the two types of particles. Considering the reuse of the separated soil, the addition of chemical substances, such as dispersants, needs to be controlled as much as possible; therefore, physical agitation was used. The suspension was constantly stirred at 100 rpm using a stirring blade (BL300, Three-One Motor) during separation. This was performed to inhibit the sedimentation of large particles.

Magnetic separation experiments were performed using a magnet bar (maximum flux density of 1 T, neodymium magnet, 25 mm in diameter, 200 mm in length, made by KTT) at an inflow velocity of 20 cm/s. During the magnetic separation experiment, the soil suspension was stirred at 100 rpm using a stirring blade.

3.2. Post-separation Analysis

After separation, the soil suspension was filtered by suction t hrough a membrane filter (pore size 1.2 µm). The filtered soil was dried, weighed, and the volume magnetic susceptibility was measured on a magnetic balance (Magnetic Susceptibility Balance, MSB Auto, Sherwood). From the measured magnetic susceptibility, we estimated the percentages of vermiculite and kaolinite in the soil. First, five types of vermiculite and kaolinite were prepared at predetermined ratios. The magnetic susceptibility of the soil samples at each ratio was measured and a calibration curve was obtained. The percentages of vermiculite and kaolinite were determined based on this calibration curve. The particle size distributions of the particles before and after separation were measured using a laser diffraction/scattering particle size distribution analyzer (LA-920, Horiba, Ltd.).

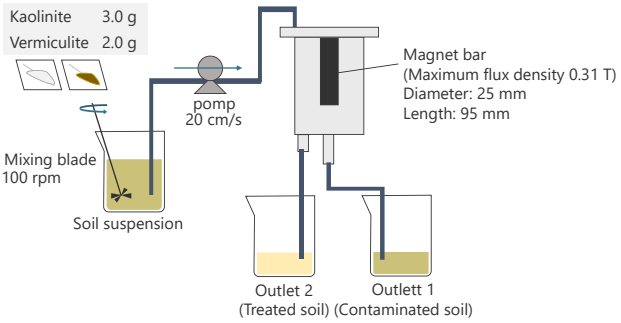

Fig. 4. Schematic view of lab-scale system of cyclone-type magnetic separation.

4. RESULTS AND DISCUSSION

4.1. Weight and Composition of Soil After Separation

Three types of soils were evaluated in this magnetic separation experiment: soil discharged from the center outlet of the device (Outlet 1 in Fig. 4), soil discharged from the side outlet of the device (Outlet 2 in Fig. 4), and soil remaining in the device after the separation experiment. The weight of the soil before and after magnetic separation and its composition are shown in Fig. 5 (a) and (b). In Fig. 5(a), the residual soil was 2.58 g, indicating that approximately half of the soil remains is a structural problem, and a slope on the underside of the apparatus would allow the residual to flow out to Outlet 1. As shown in Fig. 5(b), there was no significant difference between the soil composition of the central runoff (Outlet 1) and the lateral runoff (Outlet 2), but the latter tended to contain slightly more 1:1 clay minerals. Furthermore, the component that remained in the vessel without being directed to the sides, “Residual,” is found to contain more 2:1 type clay minerals. Although there is a need for improved separation devices, this suggests that even the weak magnetic field of a permanent magnet may be able to guide 1:1 type clay minerals to the outside and 2:1 type clay minerals to the inside.

Next, we evaluated the percentage of vermiculite and kaolinite present in the total residual and central runoff (Outlet 1) as “soils to be stored,” and in the lateral runoff (Outlet 2) as “soils to be reused.” Percentages of vermiculite and kaolinite after the separation experiment are shown in Fig. 6. The percentage of vermiculite in the soil to be stored was 41.7%, which was an increase compared to that before separation. The percentage of vermiculite in the soil to be reused was 34.0%, which was

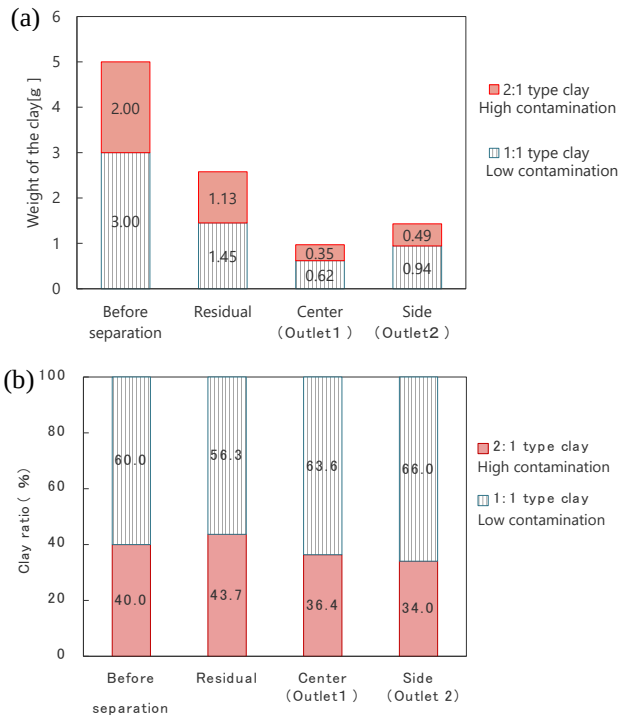

Fig. 5. (a) Weight of each clay mineral and (b) clay ratio before and after magnetic separation.

a decrease compared to that before separation. The increase and decrease in the percentage of vermiculite seem small, but indicate that the cyclone-type magnetic separator could be used to separate 2:1 type clay minerals from 1:1 type clay minerals, even with the small magnetic field strength of a permanent magnet. The maximum magnetic flux density in the magnetic separation apparatus was 0.31 T. The HGMS method for paramagnetic materials requires a magnetic field of at least 5 T of the maximum magnetic flux density of a superconducting solenoid magnet and a magnetic field gradient by placing magnetic wires within the magnetic field. However, to further improve the separation accuracy, further study of the magnetic field distribution and flow velocity distribution in the separation device and improvement of the device are necessary.

As shown in Fig. 5, the total weight of the original model soil was 5.00 g, whereas that of the treated soil (Side) was 1.43 g. Therefore, assuming that the treated soil (Side) reached a reusable radioactivity level, the volume reduction rate was expected to be approximately 30%.

4.2. Particle Size Distribution of Soil after Magnetic Separation Experiment

Fig. 7 shows the results of the particle size distributions of the soil remaining in the device, particles ejected from the center (Outlet 1), and particles ejected from the sides (Outlet 2) after the magnetic separation experiment. Most of the particles with relatively small diameters (0.2 -1.0 µm) remained in the vessel. In contrast, many of the relatively large particles (20 -100 µm in diameter) were ejected from the center. In the absence of a magnetic field, large particles would settle faster and are guided outward by centrifugal force. These results are opposite to those obtained assuming the usual principle of centrifugal separation equipment and sedimentation, suggesting the magnitude of the influence of the magnetic force.

Fig. 8 compares the particle size distributions of the remaining particles, as well as the particles from the center and side outlets, with those of vermiculite and kaolinite. Compared to the vermiculite and kaolinite particle size distributions, most of the small particles (0.2 - 1.0 µm in diameter) remaining in the vessel were considered to be kaolinite. The magnetic and centrifugal forces acting on the small particles are particularly small, and they are considered to remain in the device after diffusion. Fig. 2(a)

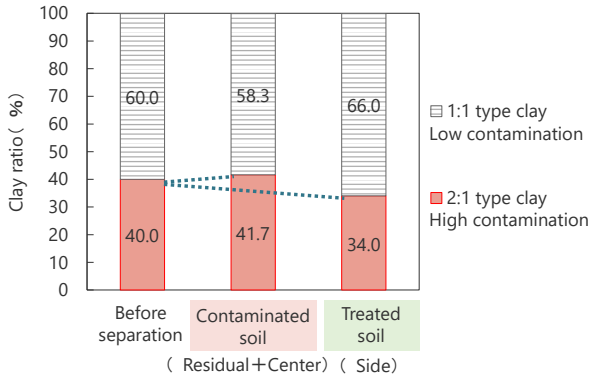

Fig. 6. Percentage of clay minerals before and after separation.

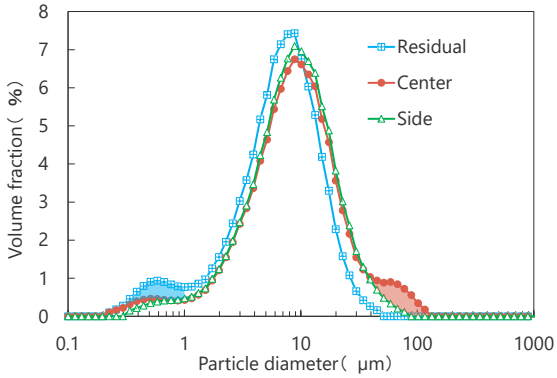

Fig.7. Particle size distribution of residual, center outlet (Outlet1), and side outlet (Outlet 2).

shows that the magnetic force acting on the 30-100 µm vermiculite is 3.75 times larger than the centrifugal force. The large contribution of the magnetic force acting on paramagnetic vermiculite is responsible for the selective separation of 2:1 type clay minerals with relatively large grain sizes.

Thus, the components of extremely large and small particles could be identified; however, for particles in the intermediate range of approximately 1-20 µm, it was not possible to identify which particles were based on the particle size distribution alone. Therefore, simultaneous analysis of the particle size and magnetic susceptibility of single particles by magnetophoresis (Kawano Laboratory Co., Ltd.) [8] was performed to identify the ratio of vermiculite to kaolinite for the residual particles and particles from the central and lateral outlets, respectively.

4.3. Estimation of Radioactivity Concentration from Separation Ratio of Soil Components

The results presented in section 4.1 only show the component ratios estimated from the magnetic susceptibility and do not consider the particle size distribution presented in Section 4.2. The component ratios are not proportional to the radioactivity concentration because the radioactivity concentration of the soil is highly dependent on its surface area. Therefore, the radioactivity concentration in the soil after magnetic separation was estimated based on the results presented in Section 4.2. The calculation assumes that all particles are spherical, and radioactive cesium is uniformly adsorbed on the spherical surface of the soil particles, depending on the adsorption capacities of vermiculite and kaolinite. Radioactivity concentration was estimated from the ratio of the surface area of the soil particles. Here, the particles discharged from Outlet 2 on the side are referred to as "purified soil," and the particles discharged from Outlet 1 on the center side and the particles remained in the device are referred to as "contaminated soil.

Equations (3) - (6) show the formulae used in the calculations.

$$X_t = X_b \times (W_t + W_c) \times \frac{A_t}{A_t + A_c} \div W_t \tag{3}$$

$$X_c = X_b \times (W_t + W_c) \times \frac{A_c}{A_t + A_c} \div W_c \tag{4}$$

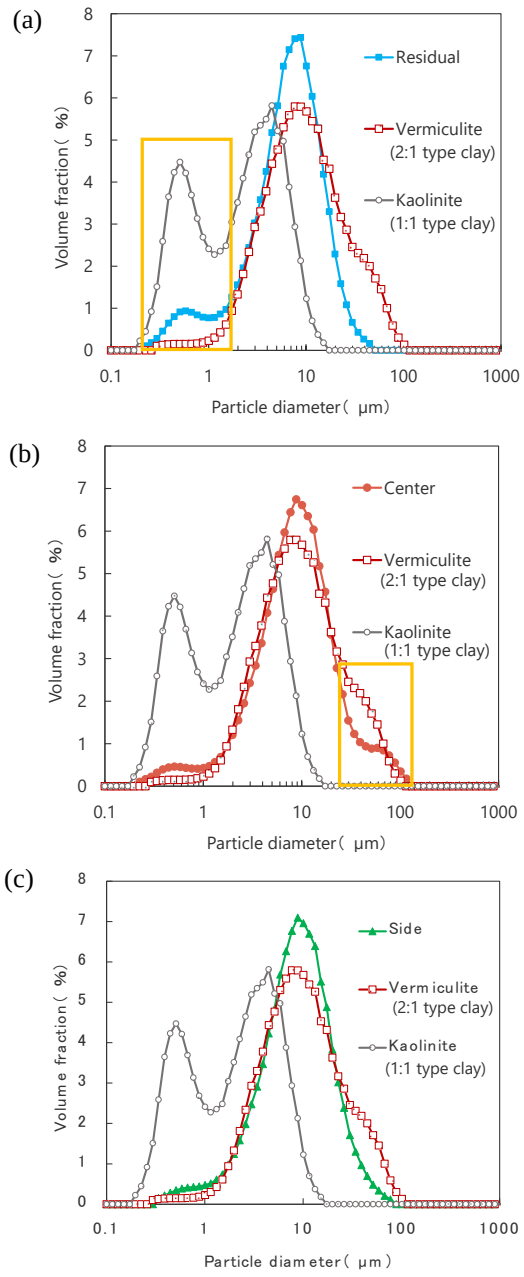

Fig. 8. Particle size distribution compared to vermiculite and kaolinite, (a) residual, (b) center (Outlet 1), and (c) side (Outlet 2).

$$A_t = \frac{W_t}{\rho} \times \sum_i \left(25 \times P_{2:1} \times \frac{p_{i,t}}{3r_{i,t}} + P_{1:1} \times \frac{p_{i,t}}{r_{i,t}/3} \right) \quad (5)$$

$$A_c = \frac{W_c}{\rho} \times \sum_i \left(25 \times P_{2:1} \times \frac{p_{i,c}}{3r_{i,c}} + P_{1:1} \times \frac{p_{i,c}}{r_{i,c}/3} \right) \quad (6)$$

Table III lists the meanings of the letters used in these equations. The subscripts "b," "t," and "c" represent the values before separation, purified (treated) soil, and contaminated soil, respectively, "2:1" represents a 2:1 type clay mineral that is paramagnetic and adsorbs a large amount of cesium, and "1:1" represents a 1:1 type clay mineral that is diamagnetic and adsorbs a small amount of cesium.

TABLE III
MEANING OF THE LETTERS IN THE FORMULA.

X	radiation concentration (Bq/kg)
W	weight of soil (kg)
ρ	volume density (kg/m^3)
A	Surface area (m^2)
p	Volume fraction (%)
r	Radius of soil particles (m)
P	Ratio of soil (-)

$X_b \times (W_t + W_c)$ in (3) represents the radioactivity of the soil before magnetic separation (Bq). The radioactivity of the purified soil was calculated by multiplying this value by the ratio $A_t / (A_t + A_c)$ of the surface area of the purified soil to the total soil. The radioactivity concentration X_t of the purified soil was calculated by dividing it by the mass W_t of the soil. The radioactivity concentration in the contaminated soil was calculated in the same manner using Equation (4).

Equation (5) calculates the surface area of the purified soil. W_t / ρ in (5) indicates the volume of purified soil, because the mass of the purified soil is divided by the density of the soil. The density of the soil was assumed to be 26,000 kg/m^3[1]. The volume of particle i was calculated by multiplying the volume of purified soil by the volume fraction of particle i. Since the volume of a soil particle is represented by $(4/3)\pi r_{i,t}^3$ and its surface area by $4\pi r_{i,t}^2$, the surface area of particle i was calculated by dividing the calculated volume of particle i by $r_{i,t}/3$. The total surface area was obtained by summing the surface area of each particle.

Considering the presence of 2:1 and 1:1 type clay minerals in the soil, the proportion of 2:1 type clay minerals, $P_{2:1}$, was multiplied by the first term in sigma and the proportion of 1:1 type clay minerals, $P_{1:1}$, by the second term in sigma. The percentages of 2:1 and 1:1 type clay minerals are based on the particle size distribution in Section 5.2, and the particle size and magnetic susceptibility of single particles measured by magnetophoresis, as shown in Table IV.

TABLE IV
PERCENTAGE OF SOIL USED FOR ESTIMATION OF RADIOACTIVITY CONCENTRATION.

	Diameter of soil particles [μm]	Ratio of 2:1 type clay [%]	Ratio of 1:1 type clay [%]
A	0.02~0.88	0	100
B	1.01~13.2	$P_{B2:1}$	100-$P_{B2:1}$
Residual		47.8	52.2
Center		34.2	65.8
Side		26.8	73.2
C	15.1~2000	100	0

Based on the particle size distribution of vermiculite and kaolinite shown in Fig. 9, area A was considered to have 0% vermiculite and 100% kaolinite, area B was considered to have $P_{B2:1}$% vermiculite and 100 - $P_{B2:1}$% kaolinite, and area C was considered to have 100% vermiculite and 0% kaolinite. Thus, the radioactivity concentration was calculated from the surface area for each particle size distribution (0.02-2000 µm). Here, $P_{B2:1}$ is the percentage of 2:1 type clay minerals present in area B. The distribution of the relationship between particle size and volumetric magnetic susceptibility of soil particles was obtained by magnetophoretic method, and the percentage of 2:1 type clay mineral present was calculated from the obtained distribution for residual, central discharge, and lateral discharge. The surface area of the contaminated soil was calculated in the same manner using Equation (4).

Here, 25 is multiplied by the surface area of 2:1 type clay minerals in (5) and (6). Akemoto et al. [9] conducted cesium adsorption experiments on vermiculite and kaolinite particles and reported that at pH 6, vermiculite adsorbed approximately 25 times more radioactive cesium than kaolinite. Because distilled water was used as the medium in this study and the pH was neutral, the surface area of 2:1 type clay minerals was assumed to be 25 times larger than that of 1:1 type clay minerals.

Fig. 10 shows a flowchart of the method used to calculate the radioactivity concentration of the simulated soils based on the ratio and particle size distribution for 2:1 and 1:1 type clay minerals, respectively. First, the particle size distribution of each component in each fractionated soil sample was determined based on the particle size distribution and magnetophoresis of the central, side, and residual particles. Based on this, the surface area of each of the 2:1 and 1:1 type clay minerals in each soil fraction was determined, and the dose for each soil was calculated from the surface area, assuming that Cs is adsorbed 25 times more on vermiculite than on kaolinite.

Fig. 11 shows the estimated radioactivity concentration. The radioactivity concentration of the soil before magnetic separation was set to 12,000 Bq/kg, which is the lowest radioactivity concentration in the soil that requires advanced treatment at intermediate storage facilities [10]. The radioactivity concentration of the soil discharged from the flanks was 8,499 Bq/kg, a 29% reduction in radioactivity concentration compared to that before magnetic separation. As described in Section 4.2, the radioactivity concentration of the soil particles discharged

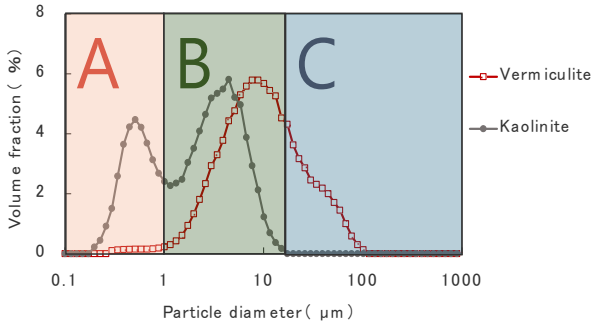

Fig. 9. Percentage of soil used for estimation of radioactivity concentration.

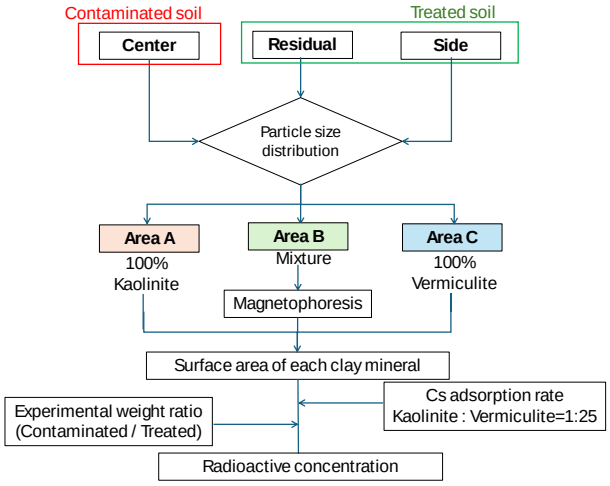

Fig. 10. Area of percentage of soil used for estimation of radioactivity concentration.

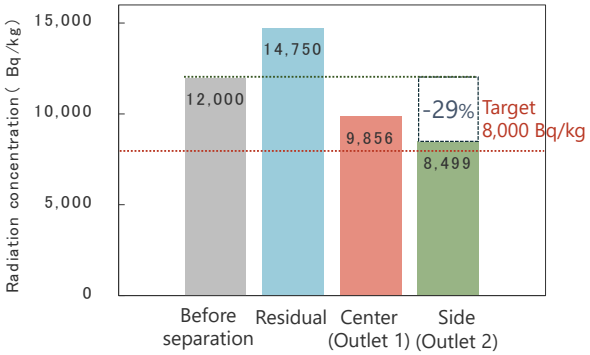

Fig. 11. Estimation of radioactivity concentration after cyclone-type magnetic separation.

from side Outlet 2 was reduced because of the presence of many small particles with a large surface area in the residue and large vermiculite particles discharged from center Outlet 1.

The radioactivity concentration calculated here is only an estimate based on the specific surface area of the separated particles and the results of existing studies on the cesium adsorption rate of two types of clay minerals. Therefore, the calculated results have limitations in terms of the expected actual effects. For example, actual clay mineral particles are plate-like rather than spherical and are non-uniformly adsorbed by crystalline planes or adsorbed between layers. It must be noted that our calculations contain large errors in this respect.

In addition, particle aggregation in actual soil must also be considered. We previously conducted a separation experiment with HGMS using contaminated soil in Fukushima Prefecture [11]. These results were considerably lower than the separation rate in the simulated experiment using a mixture of vermiculite and kaolinite. This may be because the particles aggregated with each other owing to the organic matter in the soil. This means that proper pretreatment for soil dispersion is necessary for the application of this method to real soils. It must also be considered that actual soils contain a variety of substances, not only the pure minerals used as model substances in this study.

The actual applicability to the removed soil must be verified by the separation of the contaminated soil and subsequent measurement of the radioactivity concentration. However, the new magnetic separation method we propose has demonstrated the possibility of separating paramagnetic and diamagnetic materials.

5. CONCLUSION

In this study, we proposed a cyclone-type magnetic separation technique as one of the physical volume reduction methods to reduce the final disposal volume of contaminated soil generated after the Fukushima nuclear power plant accident. The results of the calculation of the magnetic force acting on the particles indicated the possibility of separating 1:1 type clay minerals with low cesium adsorption and 2:1 type clay minerals with high cesium adsorption. Based on these results, lab-scale magnetic separation experiments were conducted using a magnet bar. The results showed that the proportion of 2:1 type clay minerals increased in the storage portion, while the proportion of 2:1 type clay minerals decreased in the reused portion. Even with a weak magnetic field strength of approximately that of the permanent magnet used in this study (maximum flux density of 0.31 T in the flow channel), we considered that a cyclone-type magnetic separation device could be expected to separate 2:1 and 1:1 type clay minerals.

To improve the separation accuracy further, we will consider using a superconducting bulk magnet as the magnetic field source and increasing the magnetic field gradient by wrapping a ferromagnetic mesh around the magnet. In addition, it is necessary to consider the shape of the lower part of the separation device and the inlet flow velocity to reduce the number of particles remaining in the device. The use of well-established techniques for typical dry or wet cyclones should also be considered. For example, it may be effective to consider using conical rather than cylindrical vessels, or using a method where the direction of flow is from bottom to top rather than from top to bottom. As the advantage of this method is maintenance-free continuous separation, the presence of residual particles in the equipment is a major challenge. These improvements in equipment geometry and flow are expected to effectively reduce the amount of residual particles.

If this method is established and improved for practical use, it will be possible to separate particles from a mixture of two types of materials, and it may be widely applied not only to removed soil but also in the field of resource recycling.

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