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Development of volume reduction method of cesium contaminated soil with magnetic separation

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Abstract

In this study, we developed a new volume reduction technique for cesium contaminated soil by magnetic separation. Cs in soil is mainly adsorbed on clay which is the smallest particle constituent in the soil, especially on paramagnetic 2:1 type clay minerals which strongly adsorb and fix Cs. Thus selective separation of 2:1 type clay with a superconducting magnet could enable to reduce the volume of Cs contaminated soil. The 2:1 type clay particles exist in various particle sizes in the soil, which leads that magnetic force and Cs adsorption quantity depend on their particle size. Accordingly, we examined magnetic separation conditions for efficient separation of 2:1 type clay considering their particle size distribution. First, the separation rate of 2:1 type clay for each particle size was calculated by particle trajectory simulation, because magnetic separation rate largely depends on the objective size. According to the calculation, 73 and 89 % of 2:1 type clay could be separated at 2 and 7 T, respectively. Moreover we calculated dose reduction rate on the basis of the result of particle trajectory simulation. It was indicated that 17 and 51 % of dose reduction would be possible at 2 and 7 T, respectively. The difference of dose reduction rate at 2 T and 7 T was found to be separated a fine particle. It was shown that magnetic separation considering particle size distribution would contribute to the volume reduction of contaminated soil.

Keywords : Magnetic separation, Radioactive cesium, Clay minerals, Particle size distribution, Particle trajectory simulation

1. INTRODUCTION

By the Fukushima Daiichi nuclear power plant accident, a large amount of radioactive substances were emitted widely. Most emitted radioactive substances deposited in the soil and a large amount of soil was contaminated. Cesium-134 and Cesium-137 were the main substances which caused the soil contamination. Now Cs^{137} which has long half-life (30 years) is of a problem and air dose will not reduce within a short period. Government has carried out the decontamination work such as topsoil stripping. The total quantity of contaminated soil discharged through this decontamination work was estimated to be 16-22 million m^3 [1]. The government has been planned to store all discharged soil in the interim storage facilities. But it is concerned that site acquisition of facilities was difficult and furthermore it takes a long time to transport to the facilities. Accordingly, the method for volume reduction of the Cs contaminated soil has been required. The volume reduction means to separate the contaminated soil into a small quantity of high-dose soil and a large quantity of low-dose soil. This contributes to reduce the amount of contaminated soil which needs to store in the facilities.

The soil is divided into four classes by their particle size; sand, gravel, silt and clay. Radioactive Cs in the soil is usually accumulated in the silt and clay [2]. This is because the smaller particles show larger specific area and hence larger amount of Cs is adsorbed on the surface of the smaller particles. It is possible to separate a large quantity of low-dose soil by classification. However volume reduction rate is not so high in a clayey soil such as field soil.

For further volume reduction, it is necessary to process the clay fraction. We focused on paramagnetic 2:1 type clay minerals in silt and clay component. They strongly adsorb and fix a large amount of Cs which is difficult to desorb. On the other hand, it is known that silt and 1:1 type clay minerals such as quartz, feldspar and kaolinite show diamagnetism and weakly adsorb a small amount of Cs. Based on these Cs adsorption mechanisms and magnetic characteristics, 2:1 type clay which is high-dose components could be separated from other minerals magnetically. Thus, it is possible to reduce the volume of high-dose soil by selective separation of 2:1 type clay by magnetic separation.

In this study, we intended to examine magnetic separation utilizing the superconducting magnet for efficient separation of paramagnetic 2:1 type clay. It consists of the particles of which size are several hundred nanometers to several tens of micrometers.

Assuming that Cs is adsorbed on the surface of the clay minerals, the radioactivity on each particle is calculated based on the measured vermiculite particle size distribution and was shown in Fig.1.

The result showed that 2-5 μm particles account for the particularly large radioactivity and hence it is necessary to

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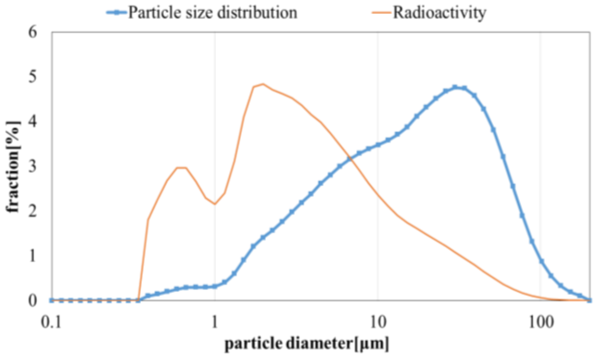

Fig. 1. Particle size distribution and radioactivity.

separate these particles for volume reduction of high-dose soil. It should be pointed that the suitable magnetic separation condition should be investigated considering particle size distribution of 2:1 type clay since magnetic force is proportional to volume of the objects. Firstly, we calculated the separation rate of the 2:1 type clay changing the particle diameter by particle trajectory simulation. Secondly, dose reduction rate by magnetic separation was calculated based on the simulation.

2. THEORY

It has been recognized that Cs ions are mainly adsorbed by silt and clay minerals whose particle diameter is smaller than 75 μm in the contaminated soil. Quartz and feldspar account for major portion of the silt. Clay minerals mainly consist of 1:1 and 2:1 type clay minerals which are different in crystal structures.

These components are different in Cs adsorption properties due to the different structure. Quartz, feldspar and 1:1 type clay adsorb Cs ions on their surfaces by the variable charge. The variable charge depends on the pH, type of cation and its concentration of solution [3]. Cs ions are weakly adsorbed when the surface of clay minerals have negative charges. Cs ions are easily desorbed by washing with the solutions containing other cations.

On the other hand, 2:1 type clay adsorbs Cs ions not only by the variable charges but also by the permanent charges originated from its structure. In the structure of 2:1 type clay, some of Si and Al ions are usually substituted by Fe and Mg ions. This causes imbalance of electrical charges that form negative charges. These negative charges are called permanent charges. Moreover, Cs ions are also fixed between the layers spatially and Cs ions which are fixed to the interlayer of 2:1 type clay are difficult to desorb. Thus, selective separation of 2:1 type clay contributes further volume reduction of Cs contaminated soil.

The magnetic properties of soil are important for the magnetic separation. The magnetic susceptibility of the soil is derived from the constituent elements. Clay minerals and the primary minerals are essentially diamagnetic because they mainly consist of silica and alumina. However 2:1 type clay contains Fe by isomorphic substitution which induces paramagnetism.

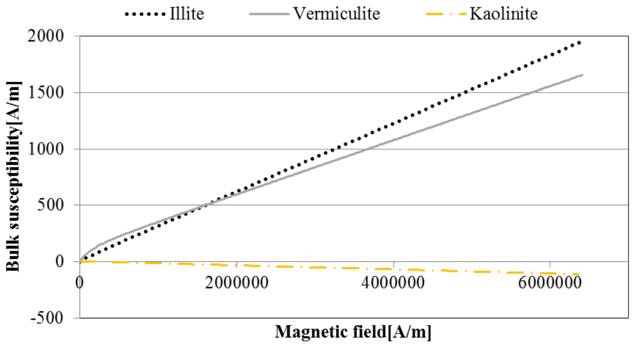

Fig. 2 Magnetization curve of the clay minerals.

TABLE I
MAGNETIC SUSCEPTIBILITY OF SILT AND CLAY.

		Magnetic susceptibility[-]
Silt	Quartz	-2.1×10^{-5}
	Feldspar	-1.3×10^{-5}
Clay	Illite	3.1×10^{-4}
	Vermiculite	2.6×10^{-4}
	Kaolinite	-1.7×10^{-5}

We measured magnetization of illite, vermiculite, kaolinite, quartz and feldspar. Illite and vermiculite are typical example of 2:1 type clay and kaolinite is 1:1 type clay. Quartz and feldspar are known to be abundant in silt.

Fig.2 shows the obtained magnetization curve and Table 1 shows the calculated magnetic susceptibility. These results show 2:1 type clay minerals are paramagnetic whereas other components are diamagnetic. This indicates 2:1 type clay could be separated magnetically.

A high magnetic field and high magnetic gradient are needed to separate the paramagnetic particles whose magnetic susceptibility is 2.6×10^{-4} and particle diameter is small. Therefore we employed a superconducting magnet for high magnetic field. Moreover the magnetic filters were also used for high magnetic gradient. The system having the magnetic filters is called High Gradient Magnetic separation (HGMS).

3. PARTICLE TRAJECTORY SIMULATION

3.1. Calculation method

As previously noted, it is possible that paramagnetic vermiculite can be separated from other soil components by HGMS [4]. However the separation rate is largely affected by the particle diameter of 2:1 type clay minerals. Accordingly it is necessary to calculate the separation rate for each particle size considering the particle size distribution for estimating the radioactivity.

Based on the observed particle size distribution which was measured by Laser diffraction-scattering type particle size distribution analyzer (LA-920, HORIBA), we conducted the particle trajectory simulation to estimate the separation rate. The objective particles were vermiculite.

TABLE II

CONDITIONS OF THE MAGNETIC FIELD AND FLUID FLOW ANALYSIS.

Analysis conditions	
Filter materials	SUS340
Wire diameter of filter	0.34 mm
Applied magnetic field	2 T, 5 T, 7 T
Flow velocity	3.0 cm/s
Viscosity coefficient	Water (1.0×10^{-3} Pa・s)

TABLE III

CONDITIONS FOR THE PARTICLE TRAJECTORY SIMULATION.

Analysis conditions	
Objective particles	Vermiculite
Magnetic susceptibility	2.6×10^{-4}
Particle size	0.1~10.1μm

The distributions of magnetic field and fluid velocity were analyzed by ANSYS 10.0 software based on finite element method (FEM). Analysis conditions are shown in Table II. The simulations were performed at 2, 5 and 7 T of maximum magnetic flux density. Drag force ($\mathbf{F}_\mathbf{D}$) and magnetic force ($\mathbf{F}_\mathbf{M}$) shown in (1) and (2) were used together with the results of magnetic field and fluid flow analysis. Here, η [Pa・s] is a viscosity of the medium, $\mathbf{v}_\mathbf{f}$ and $\mathbf{v}_\mathbf{p}$ [m/s] are the flow velocity of fluid and particles, r[m] is radius of the particles, $\mathbf{m}$ [A/m] is magnetization of the particles and $\mathbf{B}$ [T] is magnetic flux density at the position of the particles, respectively.

$$\mathbf{F}_\mathbf{D} = 6\pi\eta r_P\left(\mathbf{v}_\mathbf{f} - \mathbf{v}_\mathbf{P}\right) \tag{1}$$

$$\mathbf{F}_\mathbf{M} = V(\mathbf{m}\cdot\nabla)\mathbf{B} \tag{2}$$

Using these equations, the particle trajectory simulation was conducted by solving the motion equation of the particle shown in (3) with time evolution by Euler's method. Table III shows the analysis parameters. Particle diameters were changed from 0.1 to 10.1μm.

The area where vermiculite is captured by a magnetic wire is defined as capture radius, r_c. The separation rate with a magnetic filter, α, is defined by (4) which are written by the capture radius, r_c, wire diameter of filter, d_w and filter coefficient, f.

Then the separation rate for each particle size was calculated when 30 magnetic filters were set in the magnet. Total separation rate of vermiculite was calculated by integrating the number of the captured particles.

$$m\mathbf{a} = \mathbf{F}_\mathbf{M} + \mathbf{F}_\mathbf{D} \tag{3}$$

$$\alpha_1 = f\,\frac{r_c}{d_w} \tag{4}$$

3.2. Results and Discussions

Fig.3 shows the total separation rate with 30 magnetic filters. 73 % and 89 % of vermiculite were separated at 2 T and 7 T, respectively. The particle size distributions for captured and passed particles after magnetic separation were shown in Fig.4 and Fig.5. These results indicate that vermiculite particles from 2 to 5 μm were hardly separated at 2 T, whereas these particles could be captured at 7 T. These results corresponds to the previous study [5] in which separation rates were calculated with magnetic susceptibility of vermiculite as 7.0×10^{-4}. It was also found that we could capture vermiculite particles whose magnetic susceptibility was 2.6×10^{-4}. 2-5 μm of vermiculite particles, which have large specific surface area and the large radioactivity as shown in Fig.1, could be separated at the magnetic field at 7 T. We confirmed that high magnetic field of 7 T was needed for efficient dose reduction.

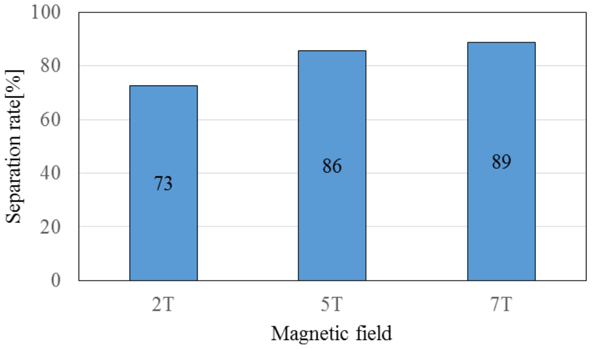

Fig. 3.Separation rate at each magnetic field.

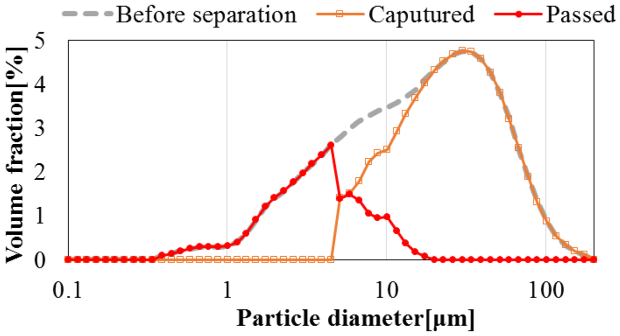

Fig .4. Calculation of particle size distribution at 2 T.

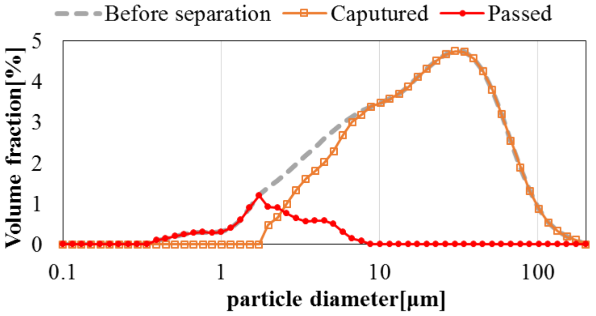

Fig. 5. Calculation of particle size distribution at 7 T.

4. CALCULATION FOR RADIOACTIVITY REDUCTION RATE

4.1. Calculation method

We calculated dose reduction rate by magnetic separation on the basis of the theory in Chapter III. We calculated under following three assumption. First, we

TABLE IV
ANALYSIS CONDITIONS FOR CALCULATION.

Analysis condition	
Magnetic separation condition	Magnetic field：2 T, 7 T Flow velocity：3cm/s Wire diameter：0.34 mm Circulation times：3 times
Soil composition	Vermiculite 25% Quartz and Kaolinite 75 %
Cs adsorption	Cs is adsorbed in proportion to the surface of vermiculite
Particle size distribution	Measured value of particle size distribution of vermiculite

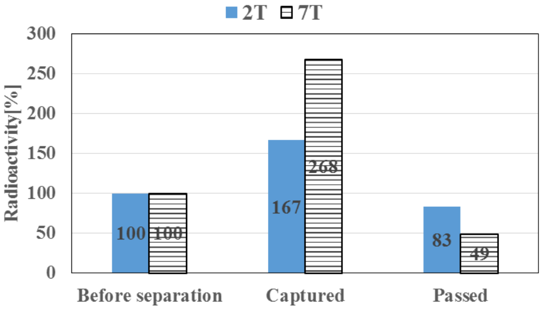

Fig. 6. Calculation of specific radioactivity after magnetic separation at 2 T and 7 T.

assumed that clay and silt were constituted by three components that are two diamagnetic materials such as quartz and 1:1 type clay minerals and one paramagnetic component of 2:1 type clay minerals such as vermiculite. Our recent investigations showed the paramagnetic vermiculite could be selectivity separated from the mixture of vermiculite and kaolinite [5]. Secondly, we supposed that diamagnetic substances were not captured by magnetic force whereas the paramagnetic particles could be captured. Thirdly, the diamagnetic substances did not adsorb Cs whereas Cs was adsorbed in proportion to the surface area of vermiculite. The table IV indicated the condition of details for the analysis.

4.2. Results and Discussion

Fig.6 shows the changes in specific radioactivity after magnetic separation under magnetic field at 2 T and 7 T. The results were normalized by the specific radioactivity before magnetic separation. From the result, magnetic separation at 2 T could reduce by 17 % in radioactivity, and at 7 T could reduce by 50 %. This difference is attributed to the captured 2~5 µm vermiculite particles which have large radioactivity.

It is also supposed that vermiculite particles smaller than 2 µm, which were hardly captured by this magnetic separation condition, has a large effect on the radioactivity of the passed soil. Therefore we need to separate vermiculite particles smaller than 2 µm for further dose reduction. We propose the multiple-step magnetic separation system in order to separate particles smaller than 2 µm. In order to separate much smaller particles, the higher magnetic field and/or the filters with smaller wire diameter would be expected to be useful in the followed separation.

5. CONCLUSION

In this study, HGMS was studied for efficient separation of 2:1 type clay minerals for volume reduction of contaminated soil. The separation rate of vermiculite was calculated by the particle trajectory simulation considering the particle size distribution.

The simulation revealed that the particle of which diameter larger than 2 µm could be separated in the magnetic field of 7 T. In addition, more than 50 % of dose reduction could be attained at 7 T. The results suggests that the magnetic separation with a superconducting magnet contributes to the volume reduction of the Cs contaminated soil.

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