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Study on magnetic separation for decontamination of cesium contaminated soil by using superconducting magnet

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The accident of Fukushima Daiichi nuclear power plant caused the diffusion of radioactive cesium over the wide area. We examined the possibility of applying magnetic separation method using the superconducting magnet, which can process a large amount of the soil in high speed, to the soil decontamination and volume reduction of the radioactive cesium contaminated soil. Clay minerals are classified as 2:1 and 1:1 types by the difference of their layer structures, and these types of minerals are respectively paramagnetic and diamagnetic including some exception. It is known that most of the radioactive cesium is strongly adsorbed on the clay, especially on 2:1 type clay minerals. It is expected that the method which can separate only 2:1 type clay minerals selectively from the mixture clay minerals can enormously contribute to the volume reduction of the contaminated soil. In this study, the components in the clay before and after separation were evaluated to estimate the magnetic separation efficiency by using X-ray diffraction. From the results, the decontamination efficiency and the volume reduction ratio were estimated in order to examine the appropriate separation conditions for the practical decontamination of the soil.

1. Introduction

By the accident of Fukushima Daiichi Nuclear Plant, many kinds of radioactive substances have been spread around the power plant. A wide range of area around the power plant is affected by the radioactive substances, hence a large amount of the contaminated soil is discharged. Because the half-life of Cs-137 is as long as 30 years among the radioactive substances, decontamination of radioactive cesium and the volume reduction of the contaminated soil are demanded. In the environment, most of cesium is adsorbed on the soil, especially clay minerals, which have the smallest particle size [1].

Some clay minerals consist of two- dimensional silica tetrahedral sheets and alumina octahedral sheets. Both sheets are piled up to form the three-dimensional structure. Clay minerals composed of tetrahedral and octahedral sheets at the rate of 2:1 or 1:1 are respectively classified 2:1 or 1:1 type clay minerals [2]. It is known that some of 2:1 type clay minerals strongly adsorb ions such as cesium in the interlayer space [3]. On the other hand, 1:1 type clay minerals have weak sorption sites on their surface where cations such as cesium ion can easily be eliminated by ion-exchange. It is also expected that cesium ions are concentrated into 2:1 type clay minerals from other clays in the environment. It is difficult to elute cesium ions from 2:1 type clay minerals without the concentrated acid or high temperature. From these findings, it is considered that the classification of clay and silt from the soil followed by selective separation of 2:1 type clay minerals contributes to decontamination and volume reduction of the cesium contaminated soil. 2:1 type clay minerals will be stored as radio isotope after the separation, because the 2:1 type clay mineral does not easily release cesium.

The 2:1 type clay minerals such as vermiculite are paramagnetic substances whereas 1:1 type clay minerals such as kaolinite are diamagnetic substances. We have attempted to apply magnetic separation with superconducting magnet to the contaminated soil using this difference of magnetic susceptibility [4].

At first, high gradient magnetic separation (HGMS) was applied to the model soil of the mixture of vermiculite and kaolinite. In order to evaluate the separation efficiency, quantitative analysis by X-ray diffraction (XRD) was performed on the soil before and after magnetic separation. Then the possibility of application of magnetic separation using the superconducting magnet was investigated. Based on this result, the magnetic separation was performed under the same condition on the schoolyard soil in Osaka prefecture in Japan. In this case, magnetic separation

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efficiency was evaluated by measuring magnetic susceptibility of soil samples captured on the magnetic filter (hereafter referred to as captured) or passed though the magnetic filter (hereafter referred to as passed).

2. Magnetic separation method

In HGMS, the ferromagnetic magnetic filter is placed in the magnetic field, in order to apply a strong magnetic force on the object. As the ferromagnetic filter is magnetized, the magnetic field gradient around it is enhanced. Enhancing magnetization generates a much stronger magnetic force which is required to separate paramagnetic substances such as 2:1 type clay minerals. In magnetic separation, particles in the fluid are mainly affected by magnetic and drag forces as shown in Eqs. (1) and (2). Gravitational force is negligible compared with these two forces under wet conditions.

$$\mathbf{F}_M = V_p(\mathbf{M} \cdot \nabla)\mathbf{H} \tag{1}$$

$$\mathbf{F_D} = 6\pi\eta r_p(\mathbf{V_f} - \mathbf{V_p}) \tag{2}$$

Here, $\mathbf{F_M}$ (N) and $\mathbf{F_D}$ (N) indicates magnetic and drag forces. V_p (m^3), $\mathbf{M}$ (Wb/m^2), $\mathbf{H}$ (A/m), η (Pa s), r_p (m), $\mathbf{v_f}$ (m/s) and $\mathbf{v_p}$ (m/s) indicates particle volume, magnetization, magnetic field strength, solvent viscosity, particle radius, solvent velocity, and particle velocity, respectively. Eq. (3) is acquired when one-dimensional approximation is applied to Eq. (1).

$$F_M = \frac{\chi}{v_0} V_p B_{ex} \frac{dB}{dx} \tag{3}$$

Here, χ [–], μ_0 (Wb/A·m), B_{ex} (T) and dB/dx represents the magnetic susceptibility of the particle, magnetic permeability in vacuum and magnetic flux density of the external magnetic field applied to the particle and the magnetic field gradient generated by the filter, respectively. The magnetic field gradient was approximated by the value of the saturation magnetization divided by wire diameter of the magnetic filter, because the magnetic filter reaches to the saturation magnetization. The magnetic separable condition is $F_M > F_D$ and flow velocity was determined from particle size and magnetic susceptibility of separation object substance.

3. Experimental method

3.1. Preparation of soil samples

In this study, seven model soil samples made of two kinds of the known standard clay minerals, vermiculite and kaolinite were prepared and analyzed by XRD. XRD analysis is used the most widely for qualitative and quantitative analysis of clay minerals [5]. Each clay mineral has each three-dimensional crystal structure, hence can be identified by X-ray diffraction. The intensity of diffracted X-ray from clay mineral mixture is related to the amount of clay components. Quantitative analysis can be performed by choosing typical peaks and preparing calibration curve.

Vermiculite (Nittai Corp., Japan) and synthetic kaolinite, which is reported as typical 2:1 and 1:1 type clay minerals in Fukushima prefecture were used as the standard clay minerals. Each standard clay mineral was sieved to be less than 75 μm. XRD analysis was performed on the vermiculite and kaolinite mixture at the weight ratio of 1:0, 4:1, 2:1, 1:1, 1:2 and 1:4 (100, 80, 67, 50, 33, 20 and 0% in percentage of vermiculite). Based on that, calibration curve was prepared. The ratios of peak intensities were calculated to obtain calibration curve. Based on the curve, quantitative analysis of clay mineral before and after magnetic separation was performed [6]. As a soil sample for HGMS, soil mixture of vermiculite and kaolinite mixed at the weight ratio of 1:1, and the schoolyard soil were sieved to be less than 75 μm. They were stirred enough within the distilled water at the weight solid–liquid ratio of 1:100.

3.2. Magnetic separation condition

Magnetic susceptibilities of vermiculite and the schoolyard soil were 2.8×10^{-4}(–) and 3.5×10^{-4}(–) respectively. The diameter of clay mineral is 5 μm and each flow velocity was calculated. From the calculation using Eqs. (2) and (3), magnetic separation was performed at 1.0 cm/s of flow velocity. The experimental condition of magnetic separation in this study is shown in Table 1.

The magnetic filter was placed inside of the cylindrical flow path (internal diameter: 7.9 mm). The path was set above the HTS (High temperature superconducting) bulk magnet (size: ϕ = 60 mm × 20 mm, the maximum surface magnetic flux density: 3.5 T). The suspension whose flow velocity was controlled by the roller pump was poured to the path, and 2:1 type clay mineral was separated selectively by the magnetic separation. The photograph and outline of the magnetic separation device is shown in Fig. 1.

The ferromagnetic stainless steel mesh (SUS430) with a saturation magnetization of 1.6 T was used as magnetic filter. The mean wire diameter of the mesh fiber was 0.15 mm and 3.0 g of the mesh was packed into 8 cm of the section of the flow path. The volume packing factor was 2.5%. The magnetic flux density at the central bottom of the path was 3.0 T.

3.3. Calculation of the separation efficiency by XRD and measurement of bulk susceptibility

XRD was performed by an X-ray diffractometer (Rigaku Corp.) on soil mixtures of vermiculite and kaolinite at indicated ratios. Measurement was performed at the range of d = 1.54–17.7 Å (2θ = 5–60°). The calibration curve was prepared by plotting the ratios of two chosen typical peak intensities (height of peaks) from XRD spectra of each soil mixture.

In order to calculate the separation ratio, XRD was performed on soil samples captured by the magnetic filter. After calculating the same peak ratio, magnetic separation efficiency was evaluated, based on the calibration curve.

3.4. Evaluation of the quantitative analysis of soil mixture by comparison of bulk susceptibility

In order to verify the result of quantitative analysis, three bulk susceptibilities were measured. Bulk magnetic susceptibilities of soil mixture before and after magnetic separation were measured by magnetic balance (Sherwood scientific, Ltd.). Bulk susceptibilities for the soil mixture which simulated the ratio obtained from XRD quantitative analysis were measured. Bulk susceptibilities were calculated by law of mixture according to the components ratios obtained.

3.5. Evaluation of magnetic separation efficiency using schoolyard soil sample

In order to evaluate the magnetic separation efficiency of the schoolyard soil, bulk susceptibilities before and after separation were measured. Bulk susceptibilities of schoolyard soils were compared.

Table 1
The experiment condition of magnetic separation.

η (Pa s)	B_{ex} (T)	dB/dx (T/m)
1×10^{-3}	3	1.1×10^4