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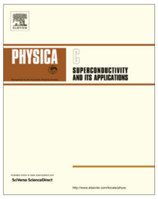

Fundamental study of cesium decontamination from soil by superconducting magnet

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A B S T R A C T

The radioactive substances have been spread out all over the surrounding area of Fukushima Daiichi Nuclear Power Plant caused by the accident in March 2011. Decontamination and volume reduction of radioactive substances, especially cesium ion, are desired issue. This study proposed a decontamination method of the soil by the magnetic separation using superconducting magnet. Cesium ion was adsorbed by Prussian blue in the potassium iodide solution. We succeeded in separating selectively the cesium ion-adsorbed Prussian blue out of the liquid phase by high gradient magnetic separation. High recovery ratio of the Prussian blue was achieved by this method.

1. Introduction

A large amount of radioactive substances have been widely scattered by the accident of Fukushima Daiichi Nuclear Plant. A wide range of area around the power plant is contaminated by the radioactive substances, hence there is a large amount of contaminated soil. Since Cs-137 has the radioactive half-life of 30 years, it is desired to remove the radioactive substances from the contaminated soil and to reduce the volume of the radioactive waste. For this purpose, we tried to develop the magnetic separation system shown in Fig. 1. In this system, we use the superconducting magnet which enables large-scale and high-throughput successive processing. For the first step, it is necessary to classify a large amount of soil and separate the clay on which the most of Cs^+ ion adsorbs. Secondly, Cs^+ ion are eluted from the clay into the solvent and collected by the adsorbent. Finally, the absorbent is recovered by the magnetic separation method.

In this study, we used the magnetic separation method by using the superconducting magnet to separate the Cs^+ ion adsorbent selectively from the soil suspension. If Cs^+ ion can be rapidly collected from a large amount of contaminated soil with this method, the radiation exposure to the operators can be reduced by the devise automated in the enclosed system. Radioactive wastes can be also collected safely in the shielded container.

It is necessary to adsorb Cs^+ ion selectively by the adsorbent in the wash fluid of the soil in order to recover the Cs^+ ion by the magnetic force. We used the potassium iodide (KI) solution as the wash fluid. Clay minerals have the high adsorption selectivity for K^+ ion followed by Cs^+ ion. It is expected that the coexisting K^+ ion, with excessive amount, desorbs Cs^+ ion from the clay minerals [1].

We used Prussian blue as the adsorbent. The Prussian blue used in this study was ammonium Prussian blue ($NH_4Fe(III)_3[Fe(II)(CN)_6]_3$) (Dainichiseika Color & Chemicals Mfg. Co., Ltd.). Prussian blue forms cubic lattice of 0.5 nm with pore where Cs^+ ion is captured selectively [2]. The paramagnetic absorbents such as Prussian blue or zeolite which can absorb Cs^+ ion are generally recovered by the coagulation settling or combination with the ferromagnetic materials [3–5], because they are not able to be recovered by a permanent magnet. A HTS (high temperature superconducting) bulk magnet enables selective collection of Cs^+ ion adsorbed paramagnetic Prussian blue by physical method of HGMS (high gradient magnetic separation).

2. Examination of magnetic separation condition

HGMS is the method that enables separating and capturing ferromagnetic or paramagnetic particles on the magnetic filters made of ferromagnetic materials by making sharp magnetic gradient around the filter. To apply this method to the separation of paramagnetic substance such as Prussian blue, highly strong magnetic field is necessary in order to increase the magnetization, hence superconducting magnet is the most suitable for such case.

Magnetic force and drag force acting on the particles in the fluid in magnetic separation method is shown in Eqs. (1) and (2). The

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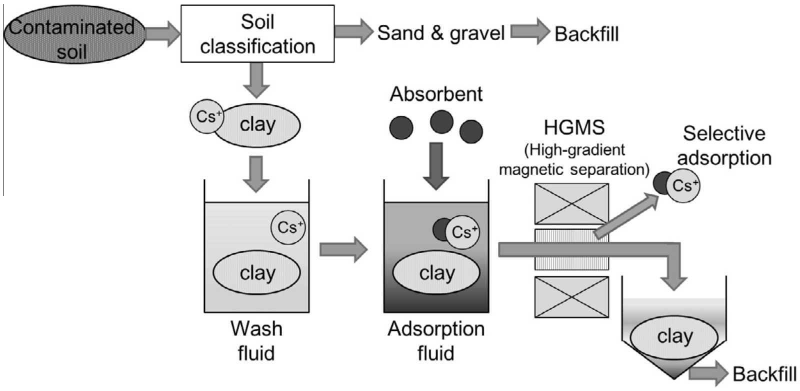

Fig. 1. Magnetic separation system.

gravitational force is negligible in the magnetic separation in fluid, since is much less than the above forces.

$$F_M = V_p(M \cdot \nabla)H \tag{1}$$

$$F_D = 6\pi\eta r_p(v_f - v_p) \tag{2}$$

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

$$F_M = \chi/\mu_0 V_p B_{ex}\, dB/dx \tag{3}$$

χ (–), μ_0 (Wb/Am), 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 the wire diameter of the magnetic filter, because the magnetic filter reaches to the saturation magnetization. The magnetic susceptibility of the Prussian blue was 9.26×10^{-5}(–). The experimental condition of magnetic separation in this study is shown in Table 1.

The condition in which magnetic separation is possible is $F_M > F_D$. It is expressed as Eq. (4) by the particle size and the fluid velocity by using Eqs. (2), (3) and Table 1.

$$3.5 \times 10^9 r_p^2 > v_f \tag{4}$$

The coefficient 3.5×10^9 is expressed by the unit of 1/ms. It was supposed that the flow velocity in which most of the particles can be separated was calculated approximately 1.4 cm/s under 3 T of the external magnetic field, based on the result that the particles observed by the microscope were more than 2 μm in diameter.

3. Experimental method

3.1. Cesium ion adsorption by Prussian blue in potassium iodide solution

We attempted to adsorb Cs$^+$ ion (Cs-133) from the KI solution which simulates the wash fluid after Cs$^+$ ion desorption from the contaminated soil. The concentrations of KI solutions were 0.01, 0.05, 0.1 and 0.5 mol/L which were prepared by adding the distilled water to KI (Kishida Chemical Co., Ltd.) and the cesium chloride (Kishida Chemical Co., Ltd.) and Prussian blue was added to the solution which resulted in the concentration of 100 ppm Cs$^+$ ion and 1000 ppm Prussian blue, respectively. This suspension was shaken at 120 rpm for 24 h so as to reach equilibrium. After filtration of the suspension, the concentration of Cs$^+$ ion was measured by ICP-MS (Inductively coupled plasma mass spectrometry) (Agilent7700, Agilent Technologies).

Table 1
The experimental condition of magnetic separation.

η (Pa s)	χ (–)	μ_0 (Wb/Am)	B_{ex} (T)	dB/dx (T/m)
1×10^{-3}	9.3×10^{-5}	$4\pi \times 10^{-7}$	3	7.1×10^4

3.2. Recovery of Prussian blue by high gradient magnet separation with HTS bulk magnet

The magnetic filter was installed inside the cylinder (internal diameter: 9 mm) which was placed vertically above the HTS bulk magnet (size: ϕ = 60 mm × 20 mm, the maximum surface magnetic flux density: 3.5 T). The suspension was pumped into the path set inside the cylinder with the control of flowing velocity. The photograph and outline of the magnetic separation device is shown in Fig. 2.

The magnetic filter was made of stainless steel wool (SUS434) with 28 μm mean diameter and its saturation magnetization was 2.0 T. The flow path was packed with 1.0 g of the wool and its filling rate was 5.7%. The magnetic flux density at the central bottom of the path was 3.0 T.

In the magnetic separation experiment, 0.1 mol/L KI solution was prepared containing 100 ppm Cs$^+$ and 1000 ppm Prussian blue and shaken for 24 h before the magnetic separation. The magnetic separation was examined at the flow rate of 1, 2, 3, 4 and 5 cm/s, based on the result of calculation of the condition in which Prussian blue particles can be recovered in high ratio. The turbidity (polystyrene turbidity) of the separated solution was measured by the turbidimeter (TR-55, Kasahara Chemical Instruments Co., Ltd.) as the indicator of separation.

4. Result and discussion

4.1. Cesium ion adsorption by Prussian blue in potassium iodide solution

Fig. 3 shows the amount of Cs$^+$ ion adsorbed on 1 g of Prussian blue in various concentration of KI solution. Generally, the amount of adsorbed Cs$^+$ ion on Prussian blue is considered to decrease by the increase of competing ion which disturbs the adsorption of Cs$^+$ ion [6]. However, in the experiment, adsorbed Cs$^+$ ion reaches the maximum at the concentration of 0.1 mol/L of KI solution, and then decreased with the increase of KI concentration up until