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Study on multi-stage magnetic separation device for paramagnetic materials operated in low magnetic fields

F. Mishima, Aoi Nagahama, N. Nomura, and S. Nishijima*

Department of Applied Nuclear Technology, Fukui University of Technology, Fukui, Japan

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Abstract

Magnetic separation technology for small paramagnetic particles has been desired for the volume reduction of contaminated soil from the Fukushima nuclear power plant accident and for the separation of scale and crud from nuclear power plants. However, the magnetic separation for paramagnetic particles requires a superconducting high gradient magnetic separation system applied, hence expanding the bore diameter of the magnets is necessary for mass processing and the initial and running costs would be enormous. The use of high magnetic fields makes safe onsite operation difficult, and there is an industrial need to increase the magnetic separation efficiency for paramagnetic particles in as low a magnetic field as possible. Therefore, we have been developing a magnetic separation system combined with a selection tube, which can separate small paramagnetic particles in a low magnetic field. In the previous technique we developed, a certain range of particle size was classified, and the classified particles were captured by magnetic separation. In this new approach, the fluid control method has been improved in order to the selectively classify particles of various diameters by using a multi-stage selection tube. The soil classification using a multi-stage selection tube was studied by calculation and experiment, and good results were obtained. In this paper, we report the effectiveness of the multi-stage selection tube was examined.

Keywords: multi-stage selection tube, magnetic separation, low magnetic field, paramagnetic particle, HGMS, soil classification

1. INTRODUCTION

A magnetic separation method is required for small-diameter paramagnetic particles, such as clay minerals in radioactively contaminated soil caused by the Fukushima nuclear power plant accident, scale and crud from power plant feedwater systems, and rare metals from urban mines. For the magnetic separation of paramagnetic particles as listed above, a superconducting high gradient magnetic separation system using a magnetic filter and a superconducting magnet as the magnetic field source has been conventionally used with an applied magnetic field of 5 T or higher (the separation effect is significant with a magnetic field of 7 T or higher). However, for use in industrial applications, it is necessary to increase the separation efficiency without using a strong magnetic field [1-4]. The initial cost of scaling up the superconducting magnet of more than 5 T required to increase throughput is enormous, making practical application difficult from an initial investment standpoint.

Hence, we are developing a selection tube magnetic separation method by applying a magnetic separation method to selection tube classification. It has been shown that magnetic separation is possible even in low magnetic fields of 0.5 to several Tesla by applying a magnetic field to small paramagnetic particles, which are suspended in a selection tube and made apparently weightless. we have confirmed improved throughput of the separation system by using a high gradient magnetic filter in the selection tube [5, 6]. The application of magnetic forces to the selection tube and the ability to separate paramagnetic materials with relatively low magnetic fields is a novelty.

Even though the magnetic field is as low as 0.5 T, when the throughput of the magnetic separation system is increased, the tube diameter becomes as large as φ500 mm to 1000 mm. Therefore, when trying to generate a 0.5 T magnetic field over a large area, we assume a superconducting magnet as the magnetic field generation source because it is difficult to use an electromagnetic magnet. It is assumed that a downward magnetic force is applied by placing the separation region of the selection tube above the maximum magnetic field region at the bore of the solenoidal superconducting magnet in the vertical direction. (We believe that the magnetic force can also control the terminal velocity of the particle and the separation speed [6]). Allowing the particles of interest to be located in the magnetic field region of the superconducting magnet (at an arbitrary location) will be a necessary technique in the future for the selection tube magnetic separation method using the superconducting magnet as the magnetic field source.

In this study, a multi-stage selection tube magnetic separation method using vermiculite minerals simulating clay minerals in radioactively contaminated soil from Fukushima was investigated and the results are reported.

2. MAGNETIC SEPARATION METHOD USING A SELECTION TUBE

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* Corresponding author: nishijima@fukui-ut.ac.jp

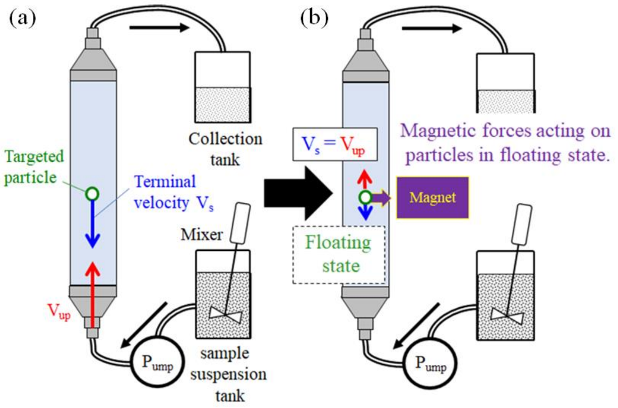

Fig. 1. Concept illustration of magnetic selection tube. (a) A selection tube in which particles are suspended by an upward flow from the bottom against the terminal velocity of the particles settling in the selection tube. (b) Captures particles suspended in the selection tube by magnetic force.

We propose a magnetic separation method using a selection tube, which is a combination of a selection tube classification and magnetic separation method.

2.1. Selection Tubes

The principle of the selection tube is shown in Fig.1(a). As shown in the figure, a selection tube is a device for classifying particles according to size by utilizing the combined forces acting on the particles based on the relationship between the settling velocity of the particles suspended in the fluid and the upward velocity of the working fluid pumped from the bottom of the device.

The particles in the selection tube are subjected to a drag force, $\mathbf{F}_D$, a buoyancy force, $\mathbf{F}_\rho$, and a gravitational force, $\mathbf{F}_g$, from the fluid. The combined force $\mathbf{F}$ acting on the particles is shown in Eq. (1), and Eq. (2) shows the final settling velocity $\mathbf{v}_s$ of the particles derived from this equation when the velocity $\mathbf{v}_f$ of the fluid (working medium) is set to 0. An upward flow $\mathbf{v}_{up}$ of the same velocity as this terminal velocity at the end of sedimentation is injected from the bottom of the selection tube so that the particles become apparently weightless and are suspended in the tube. 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], $\boldsymbol{v}_f$ is the velocity of fluid [m/s], and $\boldsymbol{v}_p$ is the velocity of the particle [m/s].

$$\mathbf{F} = \frac{4}{3}\pi r^3(\rho_p - \rho_f) - 6\pi\eta r(\boldsymbol{v}_f - \boldsymbol{v}_p) \qquad (1)$$

$$\boldsymbol{v}_s = \frac{2r^2(\rho_p - \rho_f)\boldsymbol{g}}{9\eta} \qquad (2)$$

2.2. Magnetic selection tube

Magnetic selection tube is a method of capturing and separating (collecting) particles by magnetic force on suspended particles in apparent weightlessness by an upward flow in a selection tube. (Fig. 1(b)).

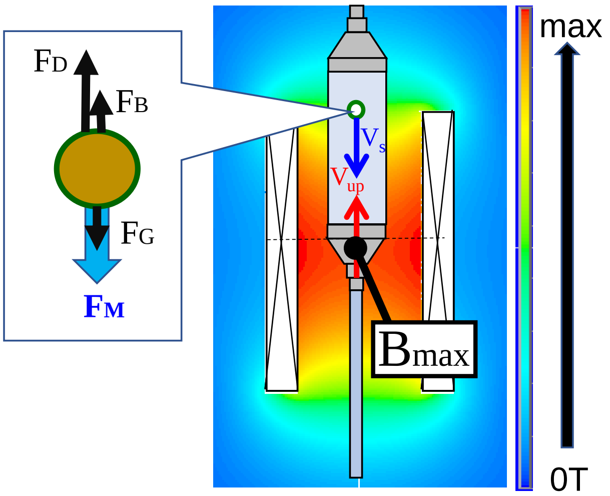

Fig. 2. Concept illustration of magnetic selection tube applied Superconducting magnet. The selection tube is located above the center of the maximum magnetic field. Magnetic flux density (B_{max}) about 0.5 to 2T.

In this separation process, the magnetic force $\mathbf{F}_M$, which attracts the particles to the magnetic field generator, and the drag force $\mathbf{F}_D$, which is the fluid resistance force, act on the particles. They are expressed by equations (3) and (4), respectively. where $\mathbf{B}$ is the external magnetic field [T], r is the particle radius, χ is the magnetic susceptibility, μ_0 is the magnetic permeability of the vacuum [H/m], η is the viscosity of the fluid [Pa·s], $\boldsymbol{v}_f$ is the fluid velocity [m/s], and $\boldsymbol{v}_p$ is the particle velocity [m/s]. Magnetic separation is possible when the magnetic force $\mathbf{F}_M$ is greater than the drag force $\mathbf{F}_D$. Since the particles are suspended in the selection tube, there is no initial drag force (technically, the drag force is generated when magnetic attraction occurs), and the force acting on the particles is dominated by the magnetic force, so particles can be captured and magnetically separated even with a weak magnetic force.

$$\mathbf{F_M} = \frac{4}{3}\pi r^3 \frac{\chi}{\mu_0}(\mathbf{B}\cdot\nabla)\mathbf{B} \qquad (3)$$

$$\mathbf{F_D} = 6\pi r\eta(v_f - v_p) \qquad (4)$$

When considering actual magnetic separation in a tube, the flow in the tube is determined by the flow line under laminar flow conditions, and the magnetic force acts spatially (three-dimensionally). In our future study, a superconducting magnet is used to apply a magnetic field (magnetic force: $\mathbf{F}_M$) that acts mainly in a downward direction to an upward direction of fluid flow (Fig.2).

3. EXAMINE THE POSITION OF PARTICLES SUSPENDED IN THE SELECTION TUBE

Previous work by our research group has confirmed that it is possible to magnetically separate only the paramagnetic glass particles in a selection tube from a mixed sample of paramagnetic black and diamagnetic yellow glass particles suspended in a selection tube by applying an open gradient field or a high gradient field. [5, 6]. In this case, a Halbach permanent magnet circuit (maximum surface flux density of 1.3 T) was used as the magnetic field source. However, the magnetic field

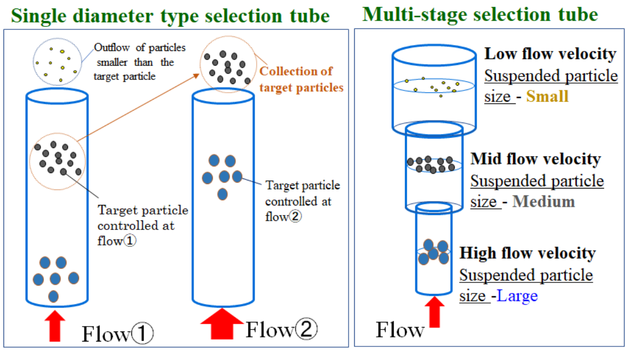

Fig. 3. Schematic illustration of the concept of a multi-stage selection tube. The single diameter type selection tube can control the particle size flowing out of the tube by the flow velocity. By changing the flow velocity several times, the target particles can be collected. By using a multi-stage selection tube with multiple flow velocities, particles of different sizes can be collected in a single operation.

generated by the permanent magnet (circuit) is very limited (a few millimeters from the magnetic surface for the permanent magnet and about 50 mm for the permanent magnet circuit).

In view of the increasing size of selection tube magnetic separators required for larger throughputs, it is desirable to use superconducting magnets, which can generate magnetic fields over a large area. When applying the magnetic field of a superconducting magnet to a selection tube. Magnetic separation using a selection tube is a technique for separating only paramagnetic particles from a group of particles of approximately the same size and density using magnetic force. We have investigated the use of a multi-stage selection tube to guide particles of the target size into the applied magnetic field area.

The target particles must be suspended in the optimal magnetic field region. However, in a conventional selection tube (cylindrical tube of the same diameter) as shown in Fig.3, the flow velocity in the vertical direction is uniform, so only particles with the same size distribution can be collected in the tube in a single operation.

To collect particles with a different size distribution, it is necessary to operate at a different flow velocity calculated from equation (2).

It is difficult to keep the region where the particles are suspended in a narrow constant region, since the position where they are suspended is determined by the position of the streamline at the initial inflow. Therefore, we proposed the use of multi-stage selection tubes with different diameters (Fig. 3).

4. MULTI-STAGE SELECTION TUBES

We have studied a multi-stage selection tube with different diameters to control the average flow velocity in each tube so as to automatically suspend particles of the target particle size at any position in the vertical direction of the tube.

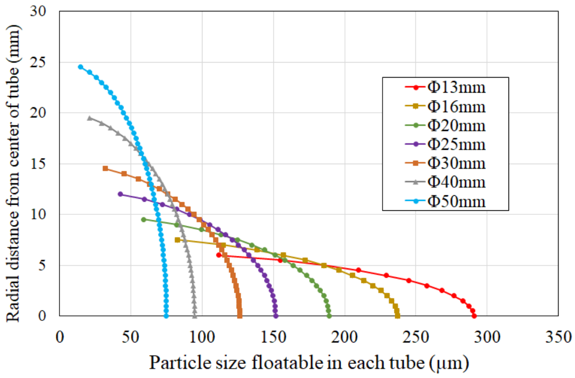

Fig. 4. calculation results for particle size capable of suspended in the 7 stages of different diameter tubes.

4.1. Examine the distribution of particles suspended in a multi-stage selection tube

In a multi-stage selection tube, the cross-sectional area of the flow channel becomes larger toward the top of the tube, and the average flow velocity and the particle diameter distributed in each tube become smaller toward the top of the tube.

The relationship between the cross-sectional area of each tube and the flow velocity determines the particles that can be suspended in each tube, so the particle size collected in each tube will also change. The working fluid in the selection tube was water. The flow in the tube was assumed to be laminar, and simulated vermiculite (density 2.5 g/cm^3) was mixed into the working fluid.

A multi-stage selection tube test apparatus consisting of 7 stages of cylindrical tubes of different diameters, each tube 20 cm long, and a total length of 170 cm including joints was investigated.

Figure 4 shows the results of calculations based on equation (2) for the particle diameters that can be suspended in each tube with different diameters and different velocity distributions. The seven-stage cylindrical tubes with different diameters selected in this study were existing commercially available clear PVC tubes with universal industrial applicability. The inner diameters were 50, 40, 30, 25, 20, 16, and 13 mm, respectively, and the flow velocity in the tubes was determined by calculation based on the initial inflow volume (240 mℓ/min). As shown in Fig. 4, when the tube diameters are close together, there is a lot of overlap in the suspended particle size distributions in the tubes, and it is desirable to reduce the number of tubes to maintain the target particle size distribution at a given point in the tubes.

From the calculation results in Fig. 5, focusing on the particle sizes suspended in the 1st, 4h, and 7th stages of all tubes, Fig. 5 shows the particle sizes floating and suspended in the tubes, approximately 100 to 300 μm in the 1st stage, 50 to 150 μm in the 4th stage, and 25 to 75 μm or smaller in the 7th stage. The overlap between the two stages is small. Next experimental study was carried out.

4.2. Classification test in a multi-stage selection tube

The multi-stage selection tube experimental apparatus had 7 stages to avoid drastic changes in flow (Fig.6). The average flow rate was set at approximately 2 mm/s (Φ50

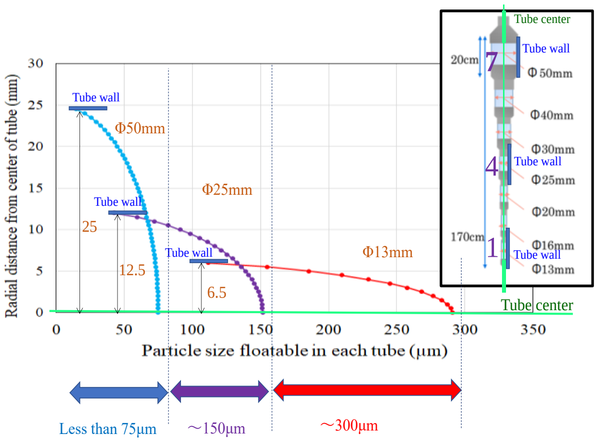

Fig. 5. Particle size of particles suspended in the 1st, 4th, and 7th stages of a multistage selection tube.

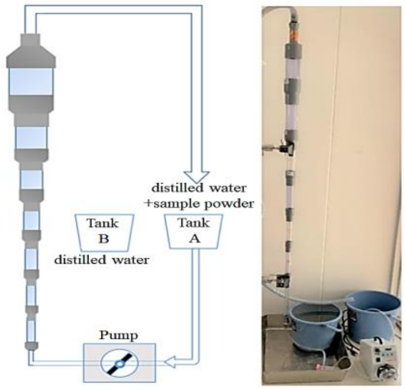

Fig. 6. Experimental setup of multi-stage (7-stage) selection tube.

mm) to 30 mm/s (Φ13 mm).The test sample was mixed with 1.0 g of ground and sieved vermiculite particles (particle size less than 300 µm) with a mesh size of 300 µm and distilled water (pH 6.5). The suspension was placed in tank A and pumped at a flow rate of 240 ml/min in a one-pass system using a metering pump from the bottom of the selection tube (tank B was drained), allowing the vermiculite particles to float in the tube and collecting particles in the 1st, 4th and 7th stages where the particle size distribution did not overlap, as shown in Fig. 5. For the parts of the recovered particles with overlapping diameters, it will be necessary to control the flow rate by subdividing the multistage diameter more finely, or to collect the particles once in an overlapped state and then classify them again by changing the flow rate.

We observed the shape and particle size of collected particles in each tube by microscope, and measured the size distribution by the analyzer (Malvern Mastersizer 3000E), and the results are shown in Fig. 7.

The microscopic observation results in Fig.7 are close to the calculated results, with particles of about 300 µm in the first stage, 100 to 150 µm in the fourth stage, and 30 to 90 µm in the seventh stage, confirming that a group of particles of a certain size accumulates in each tube.

As the results of particle size distribution analysis in Fig. 7 show, the 1st and 4th stages have characteristic distribution peaks in each tube, and the 7th stage has no overlap between the 1st and 4th stages, although the particle size distribution is wide, confirming that different groups of particles are suspended (accumulated in the tube) in each tube.

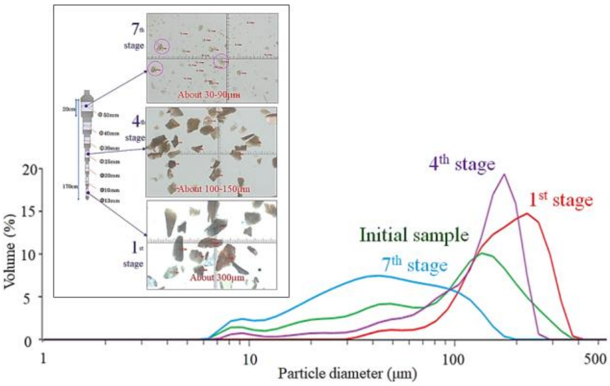

Fig. 7. Classification results using a multi-stage selection tube (1st, 4th, and 7th stages).

We believe that more consideration should be given to the results of the particle size distribution analysis, because the microscopic observation shows that the small particles were relatively spherical in shape, but the large vermiculite particles were sparsely shaped, although the collected particle diameters were close to the calculated results.

5. MAGNETIC SEPARATION TEST OF IMPROVED MULTI-STAGE SELECTION TUBE.

From the results of the previous test, a large difference in particle size distribution between the 1st and 4th stages were confirmed. Therefore, as shown in Fig. 8 (a), a selection tube with an inner diameter of 13 mm (1st stage) and 25 mm (4th stage) was connected and used in the experiment.

In previous experiments, we separated the paramagnetic particles from the diamagnetic particles of the same particle size suspended in the selection tube [5], but this experiment was performed only with the paramagnetic particles. A mixture of 1.0 g of vermiculite particles (volume magnetic susceptibility of 7.0×10^{-4}), dry classified with 105 µm and 125 µm sieves, was added to distilled water (pH 6.5). The fluid was then pumped from the bottom of the selection tube using a metering pump, and it was confirmed that particle sizes of 100 to 125 µm in diameter were suspended in the 25 mm i.d. tube at a height of 800 mm (Fig. 8 (b)).

In this process, particles near the tube wall fall due to the slow flow velocity, and as soon as they fall into the lower tube, they rise again on the upward streamline at the joint, becoming suspended and staying in the tubes, and it was observed that the target particle gradually accumulated in the tubes.

A Halbach permanent magnet circuit (maximum flux density 1.3 T) was placed on the outer wall of the tube in which vermiculite particles were suspended in the selection tube (Fig. 9(a)), and open gradient magnetic separation was performed. As shown in Fig. 9(b), after about 30 seconds, particles (particle size about 125 µm) accumulated on the

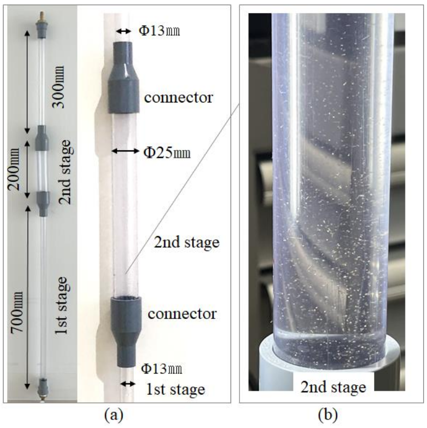

Fig. 8. (a) Photo of selection tube (2-stage type) and dimensions of tube diameter, (b) Photograph of vermiculite particles suspended in the second stage (Φ25 mm) of a two-stage selection tube (800 mm height).

inner tube wall, as shown by the dashed red circle on the magnet side.

Under the same conditions, a ferromagnetic wire filter (SUS430, wire diameter 0.3 mm, 20 mesh) was placed inside the tube so as not to interrupt the upward flow, and a magnetic field was applied to perform high gradient magnetic separation (HGMS). As shown in Fig. 9(c), almost all of the suspended particles were magnetically attracted to the magnetic filter in an instant (about 5 seconds), and we confirmed that they were magnetically separated.

This experiment showed that it was possible to accumulate (concentrate) particles of the target particle size in a suspended state at the expected position in the vertical direction of the selection tube and to separate them magnetically with a magnetic field of about 0.5T. It has also been shown that the processing speed can be improved by using the HGMS.

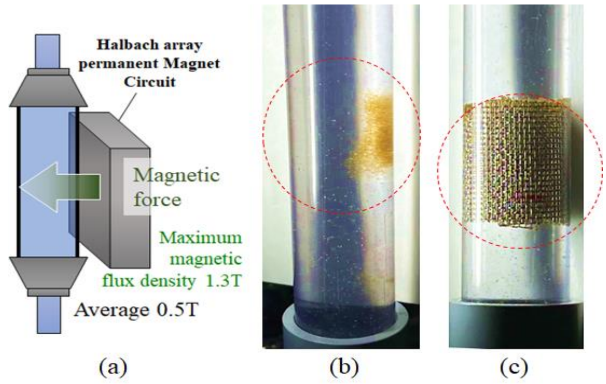

Fig. 9. (a) Schematic illustration of magnetic selection tube separation, (b) open gradient magnetic separation (OGMS), (c) high gradient magnetic separation (HGMS).

6. CONCLUSION

In this study, we investigated the classification of soil particles using a multi-stage selection tube and obtained good results that the particle size of particles suspended in the tube can be controlled according to the flow velocity distribution determined by the tube diameter.

Using our proposed method, it should be possible to concentrate ("guide") and magnetically separate particles at desired locations in the bore space of a superconducting magnet.

We plan to design and demonstrate a magnetic selection tube separator using a superconducting magnet as the magnetic field source, and also determine an efficient method for collecting the particles, which we believe is the key to making this separator practical.

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