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The development of the separation apparatus of phosphor by controlling the magnetic force

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Abstract

The phosphor wastes contain the multiple kinds of rare-earth phosphors with high market value. Because of increasing demand for rare-earth, the technique to recover and reuse the rare-earth in the phosphor wastes is required.

In this study, we focused on the difference of physical property such as magnetic susceptibility and density for each type of phosphor and tried to separate and recover the phosphors by using the magnetic separation technique utilizing the difference of the traction force to the magnet acting on the particles. Magneto-Archimedes method is the separation technique utilizing the difference of magnetic susceptibility and density. We developed the magnetic separation apparatus by applying this technique.

To develop the practical separation apparatus, the continuous process is required. Hence the fundamental experiment utilizing High Temperature Superconducting (HTS) Bulk Magnet which can generate the strong magnetic force was conducted. As a result, we succeeded the continuous separation of the phosphor wastes.

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1. Introduction

The phosphor wastes are the mixtures of the phosphors including rare earth. It is required to separate and reuse phosphors including the rare earth by the method with low cost and low environmental burden, because the cost of phosphor is rising sharply with increase of domestic demand. We proposed the magnetic separation method focusing

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attention on the difference of magnetic susceptibility, which is one of the physical properties, by the type of phosphors. Table 1 shows the types and physical properties of the phosphors used in this study. Hereafter, the name of substance was referred in abbreviation. The phosphor wastes contain a large amount of HP other than rare earth, which makes it difficult to reuse the rare earth phosphor.

<table>
<thead>
<tr>
<th>Type of fluorescent lamp</th>
<th>Typical composition</th>
<th>magnetic susceptibility (× 10⁻⁵)</th>
<th>density (g/cm³)</th>
<th>Abbr.</th>
<th>Emission color</th>
</tr>
</thead>
<tbody>
<tr>
<td>Three-wavelength type</td>
<td>Y₂O₃:Eu</td>
<td>6.97</td>
<td>5.1</td>
<td>YOX</td>
<td>Red</td>
</tr>
<tr>
<td></td>
<td>LaPO₄:Ce, Tb</td>
<td>1.33×10⁻³</td>
<td>5.2</td>
<td>LAP</td>
<td>Green</td>
</tr>
<tr>
<td></td>
<td>BaMgAl₁₀O₁₇:Eu</td>
<td>1.37×10⁻⁴</td>
<td>3.8</td>
<td>BAM</td>
<td>Blue</td>
</tr>
<tr>
<td>General color type</td>
<td>3Ca₃(PO₄)₂•Ca(F, Cl)₂•Sb, Mn</td>
<td>7.77×10⁻⁵</td>
<td>3</td>
<td>HP</td>
<td>White</td>
</tr>
</tbody>
</table>

The magnetic susceptibility of LAP and BAM is relatively large, and they can be recovered by capturing to the filter using High Gradient Magnetic Separation (HGMS) by adjusting susceptibility of the medium. On the other hand, it is difficult to separate YOX and HP by HGMS because the difference of susceptibility of YOX and HP is small. Thus, we used the magneto-Archimedes method utilizing the difference of density in addition to that of the susceptibility. In this paper, we report separation process of YOX and HP using magneto-Archimedes method.

In our previous study [1], we indicated the separation possibility of the simulated sample of the phosphor wastes using by magneto-Archimedes method. In this study, we considered the separability of the actual sample of the phosphor wastes with the goal of application to the actual recovery process. Based on the result, we designed the concept of magnetic separation apparatus which enables continuous and mass processing.

2. Theory of magneto-Archimedes method

Magneto-Archimedes method is the separation technique utilizing the difference of levitation position of each targeted particles in the medium [1-5]. The levitation position is determined by the difference of the magnetic susceptibility and density between the medium and targeted particles.

The distribution of the magnetic field is axisymmetric since the High Temperature Superconducting (HTS) bulk magnet was used as the magnetic field source in this study. The vertical component of the force \( F_{Mz} \) acting on the particles in medium is shown below:

\[
F_{Mz} = V \left[ \chi - \chi_f \left( B_r \frac{dB_z}{dz} + B_z \frac{dB_r}{dr} \right) \left( \rho_p - \rho_f \right) g \right]
\]

Defining the upward direction positive, it is possible to levitate the particles when \( F_{Mz} \) is positive. Here, \( \chi \) is the magnetic susceptibility and \( \rho \) is the density. In this regard, subscript \( p \) and \( f \) indicate particles and medium, respectively. \( \mu_0 \) is the magnetic permeability of vacuum. \( B \) is the magnitude of magnetic flux density. In this regard, subscript \( r \) and \( z \) indicate radial and vertical component, respectively. \( V \) is the volume of particles and \( g \) is the acceleration of gravity.

The radial component of magnetic force also acts both on the medium and particles. The radial component of the force \( F_{Mr} \) acting on the particles is shown below:

\[
F_{Mr} = V \chi \frac{\rho_p - \rho_f}{\mu_0} \left( B_r \frac{dB_r}{dr} + B_z \frac{dB_z}{dz} \right)
\]

The particles move by the resultant force of formula 1 and 2.

3. Particle trajectory simulation for design of continuous processing apparatus

In this study, we considered the continuous separation technique of HP and YOX, because the particles passing the filter after HGMS are mainly consists of YOX and HP. Then, we demonstrated the particle trajectory simulation for design of continuous processing apparatus.
Fig. 1 shows the schematic figure of the separation vessel. The distribution of magnetic field and of fluid velocity in this vessel was calculated using the finite element method (FEM) and the separable condition was determined.

The magnet which can generate strong magnetic force is required to levitate these particles. Hence, the HTS bulk magnet was used as the magnetic field source in this study. The HTS bulk magnet generates an inhomogeneous magnetic field which is characterized by a precipitous magnetic field gradient. HTS bulk magnet used in this study has the maximum magnetic flux density of 3.2 T above the central axis of the cryostat surface with the size of 60 mm in diameter and 20 mm in thickness. Fig. 2 shows radial and vertical distributions of the magnetic flux density.

In addition to the magnetic force from HTS bulk magnet, the drag force \( F_D \) acts on the separation targeted particles. \( F_D \) is shown by the following equation. Here, \( \eta \) is the viscosity of the fluid, \( r_p \) is the radius of the particles, \( v_f \) and \( v_p \) are the velocity of fluid and particles respectively.

\[
F_D = 6\pi \eta r_p \left( \frac{v_f - v_p}{\nu} \right) \tag{3}
\]

In the same way, the fluid analysis was performed to obtain the distribution of the fluid velocity. The inflow condition was set to 0.5 mm/sec as the initial flow rate and the outflow condition was set to zero pressure. Fig. 3 shows the distribution of fluid velocity.

The magnetic force and drag force were calculated substituting these results into the formula 1, 2 and 3. Then, trajectory of the separation targeted particles was calculated in time evolution by the Runge-Kutta method of fourth order using the motion equation shown in formula 4. Here, \( m \) is the mass of targeted particles.

\[
F_m + F_D = ma \tag{4}
\]

Fig. 4 shows the typical results of particle trajectory simulation. Referencing the previous experimental results using the simulated sample, the condition of simulation, the fluid velocity and the position and height of separation wall were determined [1]. 7.5 wt% of MnCl\(_2\) aq (magnetic susceptibility: 2.05 \( \times \) \( 10^4 \), density: 1.06 g/cm\(^3\)) was used as the medium, and the separation wall (height: 17.5 mm, thickness: 2 mm) was placed at the distance 2.5 cm from the center of magnet. The inflow velocity was set 0.5 mm/sec. Fig. 4 shows the trajectory of YOX and HP respectively. The vertical axis indicates the height from the center of magnet, and the horizontal axis indicates the radial distance. Each line represents the particle trajectory when the inflow position of the particle is respectively 3.5 cm, 3.6 cm and 3.7 cm in height above the central axis of the magnet. From the result, it was indicated that the separation of YOX and HP is possible when the inflow position is 3.5 cm in height.
4. Experiment of magneto-Archimedes method

Based on the result of previous chapter, we designed the continuous processing apparatus using magneto-Archimedes method. Fig. 5 shows the experimental system. We experimented using the designed separation vessel. As experimental condition, 7.5 wt% of MnCl₂ aq was used as the medium, and flow rate was set 5 ml/min. The suspension of the actual phosphor waste was injected from the 10 cm short of separation vessel.

UV-rays of 254 nm were irradiated to the particles which stayed inside the separation wall and exceeded the separation wall respectively, and the fluorescent color was examined. Fig. 6 shows the fluorescent photograph respectively. It is considered that the condensation of the YOX components inside of the separation wall was succeeded because the red emission was observed at right side.

Fig. 5. The experimental system of continuous separation

Fig. 6. The result of separation. Photographs of UV-rays irradiated with the particles after the separation. Left shows the particles exceeded the separation wall and right shows the particles stayed inside the separation wall.

5. Conceptual design of a large volume processing device

Based on the above results, the device to achieve a large-volume processing toward the practical use was discussed. Superconducting solenoidal magnet (maximum magnetic flux density: 10 T) was used as the source of magnetic field. The throughput is increased setting multiple separation vessels at the region of fringing magnetic field (height: 150 mm radius: 200 mm). The size of a separation unit is 6 mm in length, 100 mm in width and 150 mm in height. 54 vessels can be set on a superconducting solenoidal magnet by placing them radially at the distance of 60 mm from the center of the bore. The throughput of this device was estimated. The solid-liquid ratio of the suspension is fixed 1:50 and flow rate is 5 ml/min as similar to the previous chapter. The throughput is about 2,000 kg per year assuming that the device is continuously operated 24 hours a day for 25 days in a month.

6. Conclusion

In this study, we examined the possibility of continuous processing of the phosphor wastes using by the particle trajectory simulation. Based on the result, we designed the continuous processing apparatus and performed the separation experiment. As a result, the condensation of YOX component which is one of the rare-earth was succeeded. Based on the result, we conducted the conceptual design of large-volume processing apparatus toward the practical use. We are planning to conduct more detailed apparatus design in the future.

References