



Improvement in rare earth element recovery from waste trichromatic phosphors by mechanical activation



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ARTICLE INFO

Article history:

Received 27 December 2016

Received in revised form

20 February 2017

Accepted 13 March 2017

Available online 21 March 2017

Keywords:

Mechanical activation

Rare earth elements

Waste trichromatic phosphors

E-waste

Recovery

ABSTRACT

This research introduced a pretreatment process –mechanical activation– for extracting rare earth elements (REE) from waste trichromatic phosphors separated from e-waste. The effects of mechanical activation on the physicochemical properties of waste trichromatic phosphors were investigated using multiple methods by scanning electron microscopy (SEM), Brunauer-Emmett-Teller measurement (BET), particle size analysis, and X-ray diffraction (XRD). The results show that mechanical activation has noticeable effects on the microstructure of waste trichromatic phosphors. Increasing the rotational speed could cause the changes of S_A , structural defects, and breakage of the crystalline network, which could significantly improve in REE extraction rates. Specifically, changes in the crystalline degree of disorder caused by mechanical activation played a crucial role in enhancing the REE leachability from waste trichromatic phosphors. In addition, the sulfuric acid leaching behaviors of the activated waste trichromatic phosphors were studied by varying several parameters, including leaching time, liquid-to-solid ratio, acid concentration, heating temperature, and various activation conditions: Y, Eu, and Ce recovery rates reached approximately 96.3%, 91.1%, and 77.3%, respectively, from waste trichromatic phosphors activated for 60 min at the rotational speed of 550 rpm (by ball mill) compared with 46.7%, 42.3%, and 31.2%, respectively, from the raw sample. The method is promising to be put into practice to promote the recycling REE from various waste electric and electronic products.

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1. Introduction

Rare-earth elements (REE) are extremely valuable because of their requiring tremendous processing/refining and their critical role in electronics and other applications (Binnemans et al., 2013; Han et al., 2014). Many of these elements are essential in advanced technologies and used in phosphors, lasers, magnets, batteries, high temperature superconducting magnetic refrigeration, hydrogen transport, and more (Song and Li, 2012; Yang et al., 2012; Das and Das, 2013). Guyonnet et al. (2015) performed the material flow analysis of REE in Europe. Lanthanum, one of REE, is an important element of hydrogen-absorbing material (Morais and

Ciminelli, 2004). REE are applied in phosphors as luminescent materials (Kujawski and Pospiech, 2014; Tunsu et al., 2014). The estimated average growth rate of global REE demand is approximately 8% annually (Long et al., 2012; Tan et al., 2015). About 105,000 tonnes of REE was consumed in 2011 in globe, and the consumption may rise to 200,000–240,000 tonnes by 2020. Only in China, the annual production capacity of trichromatic color phosphors reached 20,000 tonnes (Kingsnorth, 2012). At the same time, many industrialized countries are facing problems securing sustainable supplies of REE. This problem could be alleviated by recycling end-of-life products and processing the waste streams with environmental and economic benefits (Tunsu et al., 2014; Belardi et al., 2014; Binnemans and Jones, 2014).

Among obsolete products containing REE, e-waste has been attained more and more attention (Hirajima et al., 2005a; Song et al., 2013). A European research program named “Horizon

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2020" began four years ago introduced the recycling of REE and developing new materials used for REE compounds (Belardi et al., 2014). E-waste recycling of waste phosphors in waste fluorescent lamps is a good potential source of REE, due to the large amounts sold and disposed of every year, and various fluorescent lamp recycling technologies have been explored (Tan and Li, 2014). The existing processes include acid leaching (Tian et al., 2016), dense-medium centrifugation (Hirajima et al., 2005a), flotation separation (Hirajima et al., 2005b; Otsuki et al., 2008), liquid-liquid extraction (Mei et al., 2009a; 2009b; Meor, 2013), hydrometallurgy (Rabah, 2008; De Michelis et al., 2011), photocatalytic extraction (Wu et al., 2013), ionic liquid extraction (Yang et al., 2012, 2013), supercritical extraction (Shimizu et al., 2005), and alkaline roasting (Wu et al., 2014). Due to the strong chemical stability of rare earth fluorescent phosphors, critical conditions such as high temperature and complicated experimental procedures were necessarily required for recovering REE from waste fluorescent phosphors in former research. Therefore, in order to recover waste phosphors with high efficiency easily, an environmentally friendly, energy saving process is desirable to be developed. Traditional hydrometallurgy is an established and easy technique for recovering valuable metals from second resources. However, it is not feasible for waste fluorescent phosphors. The trichromatic phosphors ($\text{BaMgAl}_{10}\text{O}_{17}:\text{Eu}^{2+}$, $\text{MgAl}_{11}\text{O}_{19}:\text{Ce}^{3+},\text{Tb}^{3+}$) are much more resistant toward acid leaching, leading to the low recovery efficiency of rare earth elements (Resende and Morais, 2010). Thus, to improve its leachability, fluorescent phosphors should be pretreated to reduce its chemical stability. Mechanical activation method as a pretreatment process has attracted more and more attention for recycling second resource (Yao et al., 2013). It can trigger physicochemical changes on the solid materials such as structural defects, phase transformations and amorphization to improve their leaching activity. Thus, to improve the leachability of various metal materials, mechanical activation method is especially suitable (Yuan et al., 2013; Ou and Li, 2014; Zhang et al., 2016). However, in reaction to recycling REE from waste trichromatic phosphors, there is no systemic investigation on the effects of mechanical activation on its physicochemical properties.

In this work, the changes in physicochemical properties and leaching rates of Y, Eu, and Ce were assessed for raw waste trichromatic phosphors and for samples mechanically activated by a planetary mill. The effects of other experimental parameters, such as liquid to solid (L/S) ratio, heating temperature, leaching time, and sulfuric acid concentration, were also investigated.

2. Experimental

2.1. Materials used

Waste trichromatic phosphors were provided by Shanghai Yuelong Rare Earth New Materials CO., LTD. The powder was dried at 105 °C for 24 h. The average chemical composition of the samples was investigated with X-ray fluorescence spectroscopy (SXF-1200; Shimadzu, Tokyo, Japan) (Table 1). Deionized water and analytical-reagent-grade sulfuric acid were used as leaching reagents.

Table 1
Chemical composition of waste fluorescent powder analyzed by X-ray fluorescence (XRF).

Compound	MgO	Al ₂ O ₃	Y ₂ O ₃	CeO ₂	Eu ₂ O ₃	Tb ₄ O ₇
Wt %	14.99	22.21	56.48	2.77	3.18	0.37

2.2. Methods

2.2.1. Mechanical activation

The waste trichromatic phosphors were mechanically activated with a planetary ball mill (PM200; Retsch, Germany). During ball milling, 20 steel balls of 10 mm diameter were used in a 50 mL bowl. In each batch, 2 g of waste trichromatic phosphors were milled in ambient atmosphere at 150, 250, 350, 450, 550, and 650 rpm with duration times of 15, 30, 60, 120, 180, and 240 min. The mechanical activation was carried out at the set rotational speed. All of the samples were sealed for subsequent characterization and leaching experiments, and underwent no further treatment after the mechanical activation.

2.2.2. Characterization

The particle sizes of waste trichromatic phosphors before and after mechanical activation were analyzed with a Microtrac particle size analyzer (MT3300; Tokyo, Japan). In this research, median particle size (D_{50}) was used. Nitrogen gas adsorption instrument (Nova4000e; Quantachrome, USA) base on BET method was applied to measure the specific surface areas of the activated samples. The samples were investigated using X-ray diffraction (XRD; Rigaku, Tokyo, Japan) with Cu K α radiation, a step of 0.02°, and a recorded range for 2θ of 10°–80° ($\lambda = 1.5406 \text{ \AA}$). The crystal plane spacing was calculated by employing Scherrer's formula from the line broadening of the XRD peaks corresponding to the (211), (222), (400), (440), and (622) planes of trichromatic phosphors (compounds with Y, Eu and Ce) with a spinel structure. The degree of disorder developed during the milling process was obtained from Equation (1) (Baláz and Kammel, 1996).

$$R = 1 - \frac{\sum I_{ac}}{\sum I_{raw}} \quad (1)$$

where R is the degree of disorder, I_{raw} is the intensity of each crystal plane diffraction peak for the raw sample, and I_{ac} is the intensity for activated samples. The values of $\sum I_{ac}$ and $\sum I_{raw}$ are obtained from XRD patterns analysis using JADE 6.0 software. The microstructures of the specimens were investigated with scanning electron microscopy (SEM; S-4800; Hatachi, Tokyo, Japan).

2.2.3. Leaching in a sulfuric acid solution

Leaching experiments on the samples in solution were carried out in a closed 250-mL glass beaker in a water bath. The magnetic stirring speed was 500 rpm throughout all leaching experiments. The leaching process was undertaken with following parameters: sulfuric acid concentrations of 0.2, 0.5, 1.0, 1.5, 2.0, and 4.0 mol/L; leaching times of 1, 4, 10, 20, 30, 40, 50, 70, 90, and 120 min; leaching temperatures of 40, 50, 60, 70, 80, and 90 °C; and L/S ratios of 180, 120, 80, 40, 20, and 10 ml/g. After leaching, vacuum filtration with a 0.45 μm nylon filter membrane was used for solid-liquid separation. The REE concentrations in the sulfuric acid leaching solution after separation were quantitatively analyzed using an inductively coupled plasma atomic emission spectrometer (ICP-AES, 1CAP6300, Thermo, USA). The extraction recovery rates were calculated by Equation (2).

$$\eta\% = \frac{[RE]_r}{[RE]_o} \times 100\% \quad (2)$$

where η is the recovery rate, and $[RE]_r$ and $[RE]_o$ are the concentrations of REE in the solution and in the waste trichromatic phosphors, respectively.

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