



Leaching of rare earth elements from waste lamp phosphor mixtures by reduced alkali fusion followed by acid leaching



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ABSTRACT

Because of the complex crystal structures of the blue powder and green powder in waste rare earth phosphor powder and the relative high content of Ce and Tb, efficient extraction of rare earth elements (REEs) is difficult. The highest leaching efficiency of REEs is only about 90% with present processes, and utilization of the REEs is low. To improve the leaching efficiency of waste rare earth phosphor, a novel process of reduced alkali fusion followed by acid leaching to recover REEs from waste phosphor powder is proposed. Under the conditions of mass ratio of reductive iron powder to waste rare earth phosphor of 1:200, alkali to phosphor mass ratio of 2:1, alkali fusion temperature of 700 °C, alkali fusion time of 3 h, hydrochloric acid concentration of 3 mol L⁻¹, liquid to solid ratio (v/w) of 7.5:1, acid leaching temperature of 70 °C, and acid leaching time of 1 h, the total leaching efficiency of REEs is as high as 99.35%, and the leaching efficiencies of Ce, Eu, Tb, and Y are 98.12%, 99.93%, 99.37%, and 99.60%, respectively. The results indicate that the REEs in waste phosphor powder can be efficiently recovered by the proposed process.

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1. Introduction

Rare earth phosphor is widely used in the field of luminescent materials, such as for fluorescent lamps, semiconductor light-emitting diodes, and cathode ray tubes (Zhou, 2004). In addition, the rapid development of the electronic information industry has increased the applications of rare earth luminescent materials (Li et al., 2007). This has resulted in the production of increasing amounts of waste rare earth phosphor powder (Tao, 2011). Because it contains a large amount of rare earth elements (REEs), waste rare earth phosphor powder needs to be recycled to relieve the pressure on resources and protect the environment (Wang, 2005; Sun and Liu, 2003; Liu et al., 2006; Chen and Qin, 2002).

There are two ways of processing waste rare earth phosphor powder: chemical and physical methods. Physical methods include the flotation separation method (Hirajima et al., 2005a), wind separation method (Takahashi et al., 2011), and centrifugal separation method (Hirajima et al., 2005b). Although red–blue–green phosphor powder can be separated and partly recovered using physical methods, the recovery efficiency is low and secondary pollution can be caused by these methods.

Chemical methods are widely used because they can extract single and pure REE from waste powders (Michelis et al., 2011; Rabah, 2008; Resende and Morais, 2010). Under the conditions of reaction temperature 37 °C, liquid/solid (L/S) ratio 50:1, stirring speed 250 rpm, concentration of sulfuric acid 2 mol L⁻¹, and leaching time 8 h, Li (2010) leached Y, Ce, Tb, and Eu with efficiencies of 75.3%, 61.1%, 66.9%, and 71.5%, respectively. Yang et al. (2012) first decomposed phosphor powder by hydrochloric acid leaching under the conditions of oxidation, roasted the leached residue with sodium carbonate, and finally dissolved the roasted product in hydrochloric acid. They achieved leaching efficiencies of 99% for both Y and Eu, but the leaching efficiencies of Tb and Ce were only 55% and 37%, respectively. At present, the process of sodium hydroxide or sodium carbonate roasting and hydrochloric acid leaching (Zhang et al., 2012) is used to recover REEs from waste rare earth phosphor powder in industry, but the total leaching efficiency of REEs is less than 90%, and the leaching efficiencies of Ce and Tb are only about 70%. The reason for the low leaching efficiencies of Ce and Tb is that trivalent Ce and Tb in phosphor powder can be oxidized to tetravalent Ce and Tb during roasting, and tetravalent rare earth oxides (REOs) are more difficult to decompose in hydrochloric acid than trivalent REOs (Zeng, 2014). We found that trivalent Ce and Tb are not converted to the tetravalent ions when reductive ferrous powder is added during the roasting process.

In this paper, a novel process involving reduced alkali fusion and acid leaching is proposed to leach REEs from waste rare earth phosphor powder. The experimental conditions, such as alkali dosage, ferrous

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dosage, alkali fusion temperature and time, hydrochloric acid concentration, S/L ratio (w/v), acid leaching time, and temperature, are then determined.

2. Materials and methods

2.1. Reagents and analysis

The experimental raw materials were mainly blue and green powder (Aluminates structures of the phosphors), and a small amount of red powder purchased from Fujian Changting Jinlong Rare Earth Company (Changting City, China). The powder sample was determined to contain 6.64 wt.% REOs by oxalic acid weight method, shown as Table 1. And its REO distribution was determined by inductively coupled plasma mass spectroscopy (ICP-MS; S115, Agilent Technologies, USA), shown as Table 2. All of the chemicals used were of analytical grade, and no further purification was needed.

2.2. Experimental procedures

In a typical experiment, a certain amount of waste rare earth phosphor powder, sodium hydroxide, and reductive ferrous powder were evenly mixed. The mixture was placed in a muffle furnace and roasted (alkali fusion conditions: heating rate 50 K min^{-1} , atmosphere air, and a corundum crucible was used for the reactor). After a period of time, the product was washed with water several times, and then the slurry was filtered. Finally, the dried filter cake was added into a three-necked bottle containing a certain concentration of hydrochloric acid. The total REE content and its element distribution in the filtered and dried leaching residue were analyzed by oxalic acid weight method and ICP-MS (S115, Agilent Technologies), respectively. The experimental conditions are shown in Table 3.

3. Results and discussion

3.1. Effect of reductive iron to phosphor mass ratio

The effect of the reductive iron powder to phosphor mass ratio on the leaching of REEs from phosphor powder investigated from 0:1 to 1:50 (w/w) with the following conditions: mass ratio of alkali to phosphor powder 1:1 (w/w), alkali fusion temperature $700 \text{ }^\circ\text{C}$, alkali fusion time 3 h, hydrochloric acid concentration 3 mol L^{-1} , L/S ratio (volume of hydrochloric acid to mass of waste rare earth phosphor powder, v/w) of 7.5:1, acid leaching temperature $80 \text{ }^\circ\text{C}$, and acid leaching time 2 h. The leaching efficiencies of total REOs (TREO) and each REO in each test are shown in Fig. 1. The results show that the leaching efficiencies of the REOs reach a peak at a reductive iron powder to phosphor mass ratio of 1:200. The leaching efficiencies of total REEs, Ce, Eu, Tb, and Y are 92.85%, 84.73%, 94.54%, 91.40%, and 94.68%, respectively, for a reductive iron powder to phosphor mass ratio of 1:200. Reductive iron powder plays an important role in the process. Further increasing of the mass ratio was not that high in efficiency.

Table 1
Chemical composition of the waste phosphor.

	REOs	Al_2O_3	BaO	SrO	P_2O_5	SiO_2
Content/%	6.64	76.26	11.89	4.34	0.29	0.58

Table 2
REO distribution of the REOs in the waste phosphor.

	CeO_2	Tb_4O_7	Eu_2O_3	Y_2O_3	Pr_6O_{11}	Gd_2O_3	Nd_2O_3
Content/%	40.47	26.08	24.85	6.32	0.29	0.19	1.80

Table 3
Experimental conditions.

Investigated effect	Values
Reductive iron to phosphor mass ratio [w/w]	0:1, 1:400, 1:200, 1:100, 3:200, 1:50
Alkali to phosphor mass ratio [w/w]	1:2, 1:1, 3:2, 2:1
Alkali fusion temperature [$^\circ\text{C}$]	400, 500, 600, 700, 800
Alkali fusion time [h]	1, 2, 3, 4
Hydrochloric acid concentration [mol L^{-1}]	1, 2, 3, 4, 5
L/S ratio [v/w]	2.5:1, 5:1, 7.5:1, 10:1
Acid leaching temperature [$^\circ\text{C}$]	50, 60, 70, 80, 90
Acid leaching time [h]	0.5, 1, 1.5, 2

3.2. Effect of alkali to phosphor mass ratio

The effect of alkali to phosphor mass ratio on the leaching of REEs from phosphor powder was investigated under the following conditions: mass ratio of reductive iron powder to waste rare earth phosphor 1:200, alkali fusion temperature $700 \text{ }^\circ\text{C}$, alkali fusion time 3 h, hydrochloric acid concentration 3 mol L^{-1} , L/S ratio (v/w) 7.5:1, acid leaching temperature $80 \text{ }^\circ\text{C}$, and acid leaching time 2 h. The results are shown in Fig. 2. The leaching efficiencies of the REEs increased with increasing alkali to phosphor mass ratio. When the alkali to phosphor mass ratio was 2:1, the leaching efficiencies of total REEs, Ce, Eu, Tb, and Y are 99.37%, 98.14%, 99.98%, 99.37%, and 99.62%, respectively.

Alkali can be used to decompose the crystal structure of rare earth phosphor, which makes the non-rare-earth elements in the phosphor soluble. They can then be separated from the REEs by washing, and the REEs can be converted to REOs. As shown in Fig. 2, the leaching efficiencies of the REEs were highest when the alkali to phosphor mass ratio was 2:1.

3.3. Effect of alkali fusion temperature

The effect of alkali fusion temperature on the leaching of REEs from phosphor powder for experiments conducted under the conditions of mass ratio of reductive iron powder to waste rare earth phosphor 1:200, alkali to phosphor mass ratio 2:1, alkali fusion time 3 h, hydrochloric acid concentration 3 mol L^{-1} , L/S ratio (v/w) 7.5:1, acid leaching temperature $80 \text{ }^\circ\text{C}$, and acid leaching time 2 h is shown in Fig. 3. The results show that the leaching efficiencies of total REEs, Ce, Eu, Tb, and Y reached 99.37%, 98.14%, 99.98%, 99.37%, and 99.62%, respectively, for an alkali fusion temperature of $700 \text{ }^\circ\text{C}$. Further increase of the alkali fusion temperature had little effect on the leaching of REEs.

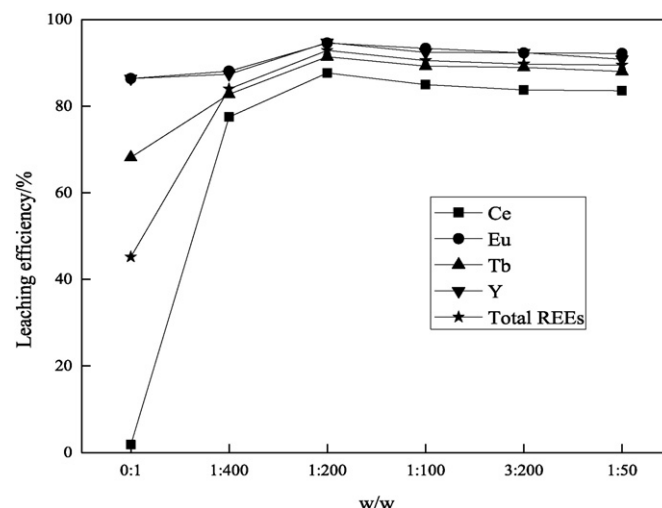


Fig. 1. Effect of reductive iron to phosphor mass ratio on the leaching of REEs.

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