



## Study on alkali mechanical activation for recovering rare earth from waste fluorescent lamps<sup>☆</sup>

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### ARTICLE INFO

#### Article history:

Received 3 April 2017

Received in revised form

30 May 2017

Accepted 31 May 2017

Available online 18 August 2017

#### Keywords:

Rare earth

Spinel structure

Recovery rate

Alkali mechanical activation

Waste phosphors

Rare earths

### ABSTRACT

Recycling of rare earth from waste fluorescent lamps is becoming increasingly important in future sustainable development. However, cerium and terbium are usually difficult to recover due to the stable spinel structure. In the research, alkali mechanical activation was innovatively used to pretreat the waste phosphors for recovering rare earths. The effect of alkali mechanical activation on the structural changes of waste phosphors was explored using X-ray diffraction (XRD) and Fourier transform infrared spectroscopy (FTIR). Meanwhile, the effects of different parameters including milling speed, activation time, and amount of alkali on the leaching rates of rare earth were investigated. The rare earth elements recovery rates are observed to increase rapidly, particularly with increases in rotational speed and activation time. Under optimal conditions, the maximum leaching rates of Ce and Tb reach 85.0% and 89.8%, respectively. And the total recovery rates of rare earth reach 95.2%. All the results indicate that alkali mechanical activation can effectively destroy the spinel structure and produce substantial physico-chemical changes. These changes lead to the easy dissolution of rare earths in acid solution, and greatly improve the leaching of cerium and terbium.

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With the increasing focus on green and low-carbon economy, fluorescent lamps have gradually replaced incandescent lamps and are widely used in lighting systems around the world, because it has a longer life, higher luminous efficiency, and less energy consumption.<sup>1–5</sup> According to statistics,<sup>6–9</sup> a large number of waste rare earth fluorescent lamps are treated as solid waste every year, which not only cause waste of rare earth resources, but also seriously pollute the environment. Therefore, the recovery of rare earth from waste fluorescent lamps has important practical significance, which can promote the sustainable development of China's rare earth resources, energy conservation, and environmental protection.

The existing processes of recycling waste phosphors are various, including acid leaching,<sup>10–14</sup> solvent extraction separation,<sup>15–17</sup>

supercritical fluid extraction,<sup>18,19</sup> alkaline fusion,<sup>20–22</sup> and mechanical activation.<sup>23,24</sup> However, these recovering methods have many shortages, such as low leaching efficiency, high energy consumption, and harsh conditions. Although it has been reported that acid leaching has the advantages of low cost, mature technology, and industrializing, the leaching rate of rare earth is lower, especially cerium and terbium. It is ascribed to the presence of aluminum-magnesium spinel structure in blue powder and green powder.<sup>25–27</sup>

Therefore, in this study, we develop a friendly alkali mechanical activation (referred to as alkali activation) as a pretreatment process technology, which can effectively destroy the spinel structure of blue powder/green powder and improve the recovery efficiency of cerium and terbium. The effects of alkali activation on the leaching rates of rare earth elements (REEs) were discussed and the optimal conditions were finding.

## 1. Experimental

### 1.1. Materials

The waste fluorescent lamp, supplied by Zhejiang Chenhui Lighting Co., Ltd., used in this research was collected from market.

<sup>☆</sup> Foundation item: Project supported by the State Key Laboratory of Advanced Processing and Recycling of Non-ferrous Metals; Lanzhou University of Technology (SKLAB02014010), the National Natural Science Foundation of China (516744130) and Natural Science Fund of Gansu Province of China (1508RJZA049).

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The chemical composition, analyzed by using X-ray fluorescence spectroscopy, is shown in Table 1. All reagents were purchased from Tianjing Chemical Works and were of analytical grade.

## 1.2. Procedure

The mixed samples of waste phosphors and alkali were mechanically activated by grinding with a planetary ball mill (QM-3SP04; Nanjing NanDa Instrument Plant; maximum speed 600 r/min). For ball milling, some ceramic balls of 5.6 mm and 9.6 mm diameter and a 50-mL bowl were used. In each batch, mixing samples and ceramic ball by 1:40 ratio were milled under an ambient atmosphere and temperature. A series of samples have been prepared at different rotational speed (250, 350, 450, and 550 r/min) and different activation time (20, 40, 60, 120, and 240 min), respectively. All the samples were subjected to leaching after the mechanical activation.

Acid leaching of inactivated and activated samples were performed by agitating 0.2 g samples into 10 mL of 2 mol/L sulfuric acid solution heated at 80 °C for 120 min. After the following leaching process, the solid–liquid separation was carried out by centrifugal machine. Concentrations of Y, Eu, Ce, and Tb in the supernatant were measured by ICP-AES analysis which were supplied by Lanzhou Institute of Chemical Physics, Chinese Academy of Sciences.

## 1.3. Analytical techniques

Compositional information had been collected, on representative aliquots of the sample, using XRF (XRF-1800, Shimadzu, Japan). The mineralogical analysis of the sample was investigated with XRD (D8 Advance, Bruker, Germany), using Cu K $\alpha$  radiation with a step size of 0.02° and a recorded range for  $2\theta$  from 5° to 80° ( $\lambda = 0.15604$  nm). FT-IR spectra were measured with a Nexus 670 infrared spectrophotometer with the KBr pellet technique. The metal concentrations in the acid leaching solution were determined by an inductively coupled plasma-atomic emission spectrometer (ICP-AES, Optima 5300DV, PerkinElmer, USA).

## 2. Results and discussion

### 2.1. Effect of alkali activation on properties

The X-ray diffraction (XRD) patterns of inactivated and activated waste phosphors are shown in Fig. 1. It is easily found that as the increased rotational speed and grinding time, the peaks intensity gradually weakens and the highest intensity diffraction peak in the inactivated sample drops to about 33% after activation at 550 r/min for 60 min. Meanwhile, the full width at half-maximum (FWHM) of peaks also gradually widens. It indicates that the crystallite size of the particles is reduced and the crystal structure of the particles is destroyed due to the impact and friction during activation.

**Table 1**  
Chemical composition of the waste phosphor analyzed by X-ray fluorescence (XRF).

Main analysis elements	Compound existence form	Content/wt%
Y	Y <sub>2</sub> O <sub>3</sub>	26.4
Eu	Eu <sub>2</sub> O <sub>3</sub>	2.18
Ce	CeO <sub>2</sub>	3.9
Tb	Tb <sub>4</sub> O <sub>7</sub>	2.2
Al	Al <sub>2</sub> O <sub>3</sub>	35.3
Ca	CaO	3.9
P	P <sub>2</sub> O <sub>5</sub>	4.5

According to the enlarged image as shown in Fig. 1(a) and (b), with the rotational speed and activation time increasing, the diffraction peaks shift to large angle, the lattice constants and cell volumes are decreased. The corresponding lattice constants and cell volume (Ce<sub>0.67</sub>Tb<sub>0.33</sub>MgAl<sub>11</sub>O<sub>19</sub>) are exhibited in Table 2. These results demonstrate that alkali activation can generate more structural changes and lattice defects.

Fig. 2 presents FTIR spectra of the alkali activation samples during different activation times (0–120 min). The results show that after 20 min of activation, new peaks located in the 1390 cm<sup>-1</sup> and 487 cm<sup>-1</sup> bands are found which are respectively corresponding to stretching vibrations of Ce–O (1390 cm<sup>-1</sup>) bond and Tb–O bond (487 cm<sup>-1</sup>). It means that the spinel structure of the green powder has begun to be destroyed during the alkali activation process. With activated time increasing to 60 min, the peaks intensity increase and the bending vibration of Ce–O bond and Tb–O bond are detected in the 1049 cm<sup>-1</sup> and 418 cm<sup>-1</sup>. It can be seen that the vibrational modes of some Ce–O and Tb–O bonds are changed, which are caused by the change of the angle of the Ce–O bond and Tb–O bond.<sup>28–30</sup> Meanwhile, the peaks intensity increases obviously, which display that the degree of structural damage is deepened. Continue to prolong activation time, although the degree of damage increases, the particles will exhibit obvious agglomeration phenomena.

These results show that the structure of the waste phosphors has been changed in the course of alkali activation, which is in agreement with the above XRD results. Meanwhile, the formation of the Ce–O bond and the Tb–O bond also demonstrated that the spinel structure has been successfully destroyed by alkali activation treatment.

### 2.2. Effect of alkali activation conditions on leaching rate

The effect of rotational speed on the extraction efficiency of REEs was investigated in the ranges of 250–550 r/min, as shown in Fig. 3(a). The leaching rates of REEs increases with the increase of rotational speed. It is clearly observed that the leaching rates of Ce and Tb are significantly elevated by ~36.9% and ~51.6%, respectively, when the rotational speed increased from 250 to 550 r/min. The results display that higher rotational speed can cause more serious lattice deformation and structural defects, which can accelerate the destruction of the crystal structure and the leaching of cerium and terbium.

Fig. 3(b) represents the leaching rates of REEs activated at different activation time. The activation time at the rotational speed of 550 r/min can also positively influence the leaching rates of REEs. Clearly, the recovery rates of REEs significantly improved with the increased activation time from 20 to 60 min. Similarly, the leaching rates of Ce and Tb have the most remarkable enhance, which increase from 40% and 33.1% to 85% and 89.8%, respectively. This also shows that the destruction of the spinel structure is exacerbated with the activation time increasing. After activation for 60 min, all leaching rates of Y, Eu, Ce, and Tb achieve the maximum. However, continue to extend activation time, the leaching efficiency is reduced due to powder agglomeration phenomenon.

Besides rotational speed and activation time, the effect of NaOH-to-waste mass ratio on the recovery rates of REEs was also studied; the results at 550 r/min and 60 min are shown in Fig. 3(c). The leaching rates of REEs are improved with the increased NaOH-to-waste mass ratio from 0:1 to 3:1. When mass ratio is 3:1, the leaching efficiency of Ce and Tb have reached to 85.0% and 89.8%. The results indicate that the more addition of alkaline can accelerate mass transfer velocity between the reactants and the reaction efficiency, thus increasing the REEs extraction efficiency. Whereas, the extraction efficiency is significantly reduced when the mass ratio of NaOH-to-waste exceeds 3:1. It is because that excess of

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