Leaching kinetic study of Y and Eu from waste phosphors using hydrochloric acid solution containing hydrogen peroxide

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Abstract: The leaching kinetics of Y and Eu from waste phosphors using a hydrochloric acid (HCl) solution containing hydrogen peroxide (H_2O_2) were investigated. Characterization of the waste phosphors was performed using XRD to ascertain the phases as $(Y_{0.95}Eu_{0.05})_2O_3$ (red phosphors), $(Ce_{0.67}Tb_{0.33})MgAl_{11}O_{19}$ (green phosphors), $(Ba_{0.9}Eu_{0.1})Mg_2Al_{16}O_{27}$ (blue phosphors), and SiO_2 (quartz). The influence of factors such as HCl concentration, addition amount of H_2O_2, temperature, and reaction time on the leaching performance of Y and Eu was investigated. The maximum leaching recoveries of Y (99.87%) and Eu (88.72%) were obtained at 4 M HCl, 0.2 cm³/g H_2O_2, 60 ℃ temperature, and 180 min of reaction time at a liquid-to-solid ratio of 7.5 cm³/g. Leaching kinetic results showed the best fit with the shrinking sphere model $(1-(1-x)^{1/3})=k_ct$, ensuring that the overall leaching process was governed by a chemical control mechanism. Activation energies of 42.35 and 33.28 kJ/mol were acquired for leaching of Y and Eu, respectively, at 40-70 ℃, which further supports the proposed chemical control leaching process.

Introduction

The demand for rare earth elements has grown owing to the numerous usages of these elements in various high-tech applications and sustainable technology (permanent magnets, nickel-metal hydride batteries, lamp phosphors) (Massari and Ruberti, 2013). Because of the scarcity of rare earth elements in the global market and limitation of minerals bearing such elements, recycling of rare earth elements from secondary sources has become increasingly important (Binnemans et al., 2013).

Phosphors are widely used in various lighting products such as fluorescent lamps, semiconductor light-emitting diodes and cathode ray tubes. Phosphors have usually been discarded as waste products from those lighting materials (Hirajima et al., 2005). Because of the presence of mercury, lighting materials such as fluorescent lamps are classified as hazardous waste (Belardi et al., 2014). After removal hazardous materials, the lamps are crushed and screened to separate glasses and phosphors. About 47 Mg glass, 2.5 Mg aluminum, and 6 Mg of non-recyclables are obtained along with each Mg of waste phosphors (Belardi et al., 2014). The obtained waste phosphors are usually either heated or washed by acetone solution to remove mercury. Owing to their high contents of rare earth elements, such as Y (26.47%), Eu (2.22%), Ce (2.61%), and Tb (1.53%) (Table 1), waste phosphors have received wide attention as urban mining resources. In 2011, about 4800 million fluorescent lamps were disposed (Zhang, 2012), and the value of rare earths contained in waste phosphors was more than 1600 million dollars (estimated according to the market price of rare earths at the close of 2016). Therefore, to lower production costs, reduce waste and prevent environmental pollution, recovery of rare earth elements from waste phosphors can be a suitable approach.

Waste phosphors generally contain a mixture of red, blue and green phosphors. The red phosphors are present as rare earth oxides and can be easily decomposed using hydrometallurgical processes.

The chemical structures of the blue and green phosphors are spinel structures (Wu and Cormack, 2003; Tang et al., 2006), which are so stable that they are difficult to destroy with inorganic acids (Luidold et al., 2012). Several researchers have reported selective leaching of Y and Eu from waste phosphors (Binnemans et al., 2013; Wu et al., 2014). Using acid leaching, purification and precipitation processes, the production of Y oxides can be obtained from waste cathode ray tubes and lamps' fluorescent powders (Innocenzi et al., 2013). Tunsu et al. (2014, 2016) investigated the effect of different agents on leaching of rare earth elements. They found that a weak nitric acid solution was the optimum agent for prioritized leaching of Y and Eu and selective separation of rare earth elements was achieved with Cyanex 923. By leaching using different acids, De Michelis et al. (2016) proved that red phosphors could be dissolved in an inorganic acid solution effectively. Using hydrochloric acid (HCl), red phosphors could be effectively dissolved by leaching (Yang et al., 2012). Via chlorination roasting followed by a water leaching process, Y and Eu can be decomposed and extracted efficiently (Yu et al., 2016).

Leaching kinetic studies of rare earth elements from secondary sources have been carried out in recent years. Yang et al. (2014) reported that leaching kinetics of rare earths from waste nickel-metal hydride batteries in HCl solution followed the chemical-reaction-controlled shrinking core model. Utilizing sulfuric acid solution, the kinetics for leaching rare earth metals from the spent nickel-metal hydride batteries also followed the chemical-reaction-controlled shrinking core model (Meshram et al., 2016). Leaching kinetic study of neodymium from scrap magnets using acetic acid followed the shrinking sphere kinetic model (Behera and Parhi, 2016). Because chemical reactions of rare earths in acid vary depending on the character of particles and the type of phases, the leaching kinetics of all the rare earths may not follow the same pattern exactly. To date, no report exists on the leaching kinetics of Y and Eu from waste phosphors in the presence of hydrogen peroxide (H_2O_2). Thus, the purpose of this study was to investigate the leaching kinetics of Y and Eu from waste phosphors using HCl solution containing H_2O_2. The effects of different variables on the leaching percentage were examined, and the kinetic model as well as the apparent activation energy were determined.

2. Materials and methods

2.1 Materials and equipment

The waste triband phosphors used in this study were mercury free and supplied by the Peiyuan Lighting Materials Co., Ltd. (Zhejiang, China). Waste lamps were crushed in an acetone solution to absorb their mercury, and then screened to separate the electrical contacts, metal ends and phosphors. Because of this, the isolation of pure phosphors was not possible, as this fraction contained many fragments of glass (SiO_2). All the chemicals used in this study were analytical-grade reagents.

An X-ray fluorescence (XRF) analysis was conducted using an Axios system (PANalytical. B.V., Co., Ltd., EA Almelo, The Netherlands), and an X-ray diffraction (XRD) analysis occurred via an RU-200B (Rigaku Co., Ltd., Tokyo, Japan). A Prodigy7 (Teledyne Leeman Labs Inc., USA) was used to conduct an inductively coupled plasma optical emission spectroscopy (ICP-OES) analysis, and a scanning electron microscope/energy dispersive spectroscopy (SEM-EDS) analysis was carried out via a Zeiss Ultra Plus SEM (Carl Zeiss AG, Germany).

2.2 Leaching of waste phosphors

Leaching experiments were carried out in three-neck round bottom flasks that were placed in a water bath (set at a desired temperature). Waste phosphors (20 g) were transferred to the flask, which contained the desired concentrations of HCl and H_2O_2 maintained at a preset temperature. The slurry was stirred with a magnetic stir bar at 600 rpm for the duration of leaching. The leaching reaction is:

$$(Y_{0.95}Eu_{0.05})_2O_3 + 6HCl = 1.9YCl_3 + 0.1EuCl_3 + 3H_2O. \tag{1}$$

The effects of acid concentration, addition amount of H_2O_2 (30%), temperature, liquid-to-solid ratio (L/S; HCl solution/waste phosphors), and reaction time on leaching efficiency were investigated. During the leaching experiments, one parameter was changed, while the others remained constant. At the end of leaching, the slurry was filtered, and the residue was washed with distilled water and dried. All the solutions generated during leaching were analyzed via ICP-OES, and residues were

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characterized using SEM-EDS and XRD. The leaching percentage was calculated using the following equation:

$$X = (a / \beta) \times 100\%,\tag{2}$$

where X is the leaching percentage of Y and Eu, a is the content of Y and Eu in the leaching solutions (ICP-OES), and β is the content of Y and Eu in the waste phosphors (XRF).

3. Results and discussion

3.1 Characterization of waste phosphors

The powder was characterized using XRD, XRF, and SEM. The XRF results indicate that the phosphors contained 26.47% Y_2O_3, 2.22% Eu_2O_3, 1.53% Tb_4O_7, and 2.58% CeO_2, along with 32.15% Al_2O_3, 18.13% SiO_2, and other trace elements such as Cu and K (Table 1). The total content of rare earth oxides was 32.83%. It was noted that the content of SiO_2 was 18.13%, which is much higher than expected for the phosphors (there is no Si in the chemical formula of phosphors). This may be because a certain amount of Si comes from the fluorescent tubes.

Table 1. Chemical composition of the actual waste phosphors

Element	Na_2O	MgO	Al_2O_3	SiO_2	SO_3	CaO	K_2O	BaO	CeO_2	CuO_3
Content (%)	1.47	2.32	32.15	18.13	0.081	2.48	1.13	3.45	2.61	0.21
Element	Eu_2O_3	Y_2O_3	Fe_2O_3	Fe_2O_3	SrO	P_2O_5	PbO	Tb_4O_7	Cl	Loss on Ignition
Content (%)	2.22	26.47	0.52	0.52	0.51	1.05	0.38	1.53	0.039	3.18

The XRD analysis of the waste phosphors is shown in Fig. 1. The primary crystalline phases of the samples were $(Y_{0.95}Eu_{0.05})_2O_3$ (red phosphor), $(Ce_{0.67}Tb_{0.33})MgAl_{11}O_{19}$ (green phosphor), $(Ba_{0.9}Eu_{0.1})Mg_2Al_{16}O_{27}$ (blue phosphor), and SiO_2 (quartz).
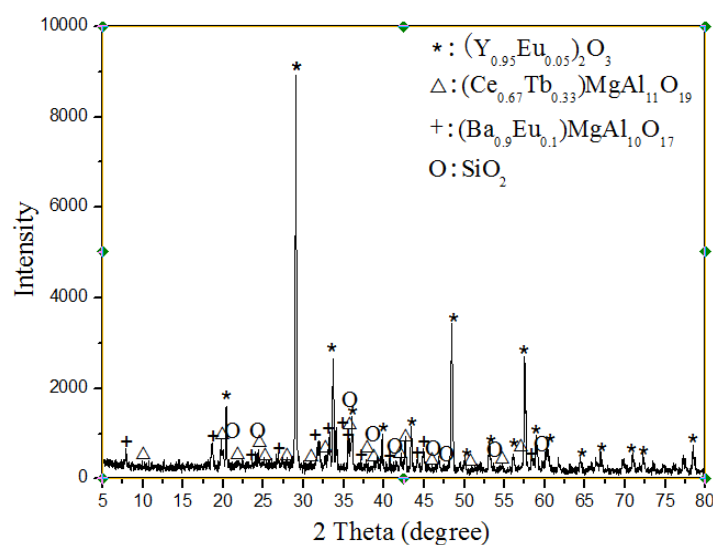

Fig. 1. XRD analysis of waste phosphors

Fig. 2 and Table 2 show the SEM-EDS analysis results for the original waste phosphors. The large granular particles in Fig. 2 are fluorescent tube fragments (point 1). The elements present in these particles are primarily Si and O (Table 2), and the average particle size of those fragments is approximately 25 μm. The primary elements of bright particles representing red phosphors (point 2) are Y and O, whereas the primary elements of both point 3 (green phosphors) and point 4 (blue phosphors) are O and Al (Table 2).
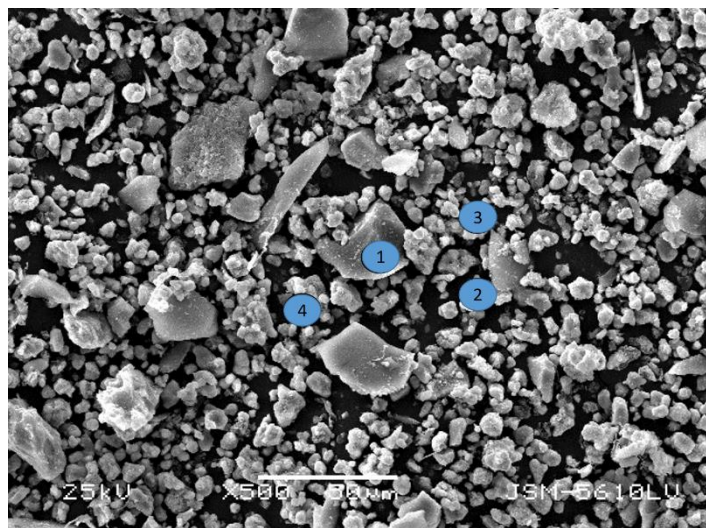
Fig. 2. SEM-EDS image of waste phosphors

Table 2. EDS analysis of waste phosphors

Elements	Content (wt%)			
	Point-1	Point-2	Point-3	Point-4
O	46.24	31.22	43.54	38.50
Mg	0.45	0.37	2.16	2.63
Al	2.91	5.09	37.21	35.05
Ca	1.03	0.74	-	0.29
Si	29.88	-	3.89	2.35
Y	-	57.22	8.15	3.42
Ba	1.48	1.38	5.46	12.41
Ce	-	0.38	4.46	-
Tb	-	-	2.65	-
Eu	-	2.79	2.62	2.66

3.2 Effect of HCl concentration on leaching percentage of Y and Eu

The experiment on the effect of HCl concentration on the leaching recovery of Y and Eu from the waste phosphors was carried out at 60 °C, 0.2 cm^3/g H_2O_2, and L/S of 10 cm^3/g for 180 min. As shown in Fig. 3, the acid concentration played an important role in the leaching process. The leaching percentage of Y and Eu increased with increasing HCl concentration up to 4 M. The rare earth elements of Y (99.84%) and Eu (88.72%) present in the red phosphors were mostly leached out. The Eu elements of the blue phosphors were very stable such that a further increase in the HCl concentration did not lead to an increase in leaching percentage of Eu. From this study, it can be ascertained that 4 M is sufficient for dissolution of both Y and Eu present in red phosphors. Hence, the optimum HCl concentration required for leaching was considered 4 M for further experiments.

3.3 Effect of addition of H_2O_2 on leaching percentage of Y and Eu

The effect of H_2O_2 concentration on the leaching percentage of Y and Eu was investigated by varying the addition of H_2O_2 (30%) from 0 to 0.4 cm^3/g under the condition of 60 °C for 180 min. As shown in Fig. 4, the leaching percentage of Y and Eu increased from 86.37 to 99.48% and from 41.29 to 88.72%, respectively, with the increase of H_2O_2 addition amount from 0 to 0.2 cm^3/g. A further increase in the H_2O_2 addition did not show any improvement in the leaching percentage of Y and Eu. Therefore, all following experiments were carried out using 0.2 cm^3/g H_2O_2. It was concluded that the addition of H_2O_2 could increase the leaching percentage of Eu significantly.
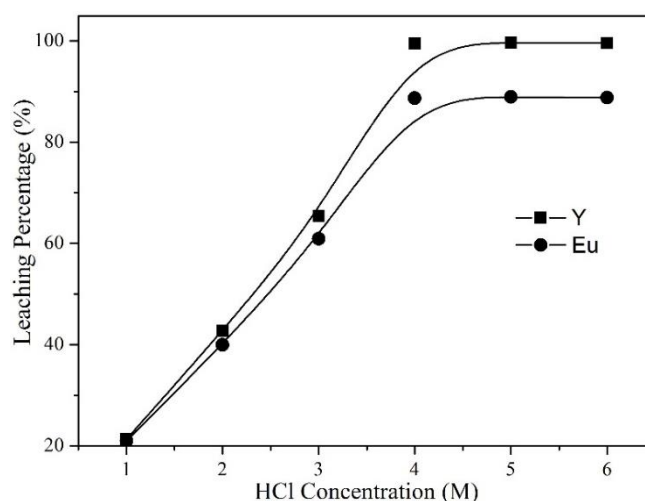
Fig. 3. Effect of HCl concentration on leaching percentage of Y and Eu
(H_2O_2: 0.2 cm³/g T: 60 ℃, t: 180 min, L/S: 10 cm³/g)
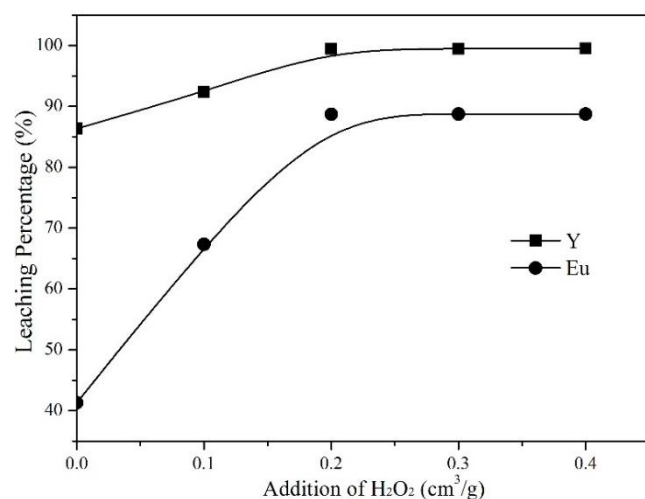
Fig. 4. Effect of addition of H_2O_2 on leaching percentage of Y and Eu
(acid: 4 M HCl, T: 60 ℃, t: 180 min, L/S: 10 cm³/g)

3.4 Effect of temperature and reaction time on leaching percentage of Y and Eu

The effect of temperature on the leaching percentage of Y and Eu was studied using 4 M HCl, 0.2 cm³/g H_2O_2, and L/S of 10 cm³/g, in the temperature range 40-70 °C for 180 min. The results indicate that high temperature promotes the decomposition of waste phosphors (Fig. 5). The leaching percentage of Y and Eu was increased from 45.21 to 99.48% and from 40.75 to 88.72%, respectively, with the increase of temperature from 40 to 60 °C. This was because leaching processes are endothermic, therefore high reaction temperatures promote dissolution of red phosphors in the acidic media. Further increase in temperature did not show any significant improvement in leaching of Y and Eu elements, and high temperature promotes volatilization of HCl, causing severe environmental pollution. Thus, to decrease energy consumption and prevent air pollution, 60 °C was chosen as the optimum temperature for leaching of Y and Eu elements.

The reaction time of leaching is another important factor that influences the leaching process. The reaction time effect was examined in the range of 40-300 min under the conditions of 60 °C, 4 M HCl, 0.2 cm³/g H_2O_2, and L/S of 10 cm³/g. As shown in Fig. 6, the leaching percentage of Y and Eu was increased from 42.23 and 37.33% to 99.48 and 88.72%, respectively, with an increase in the reaction

time from 40 min to 180 min, and thereafter the leaching percentage was constant through 300 min. Hence, 180 min was considered the optimum time of leaching
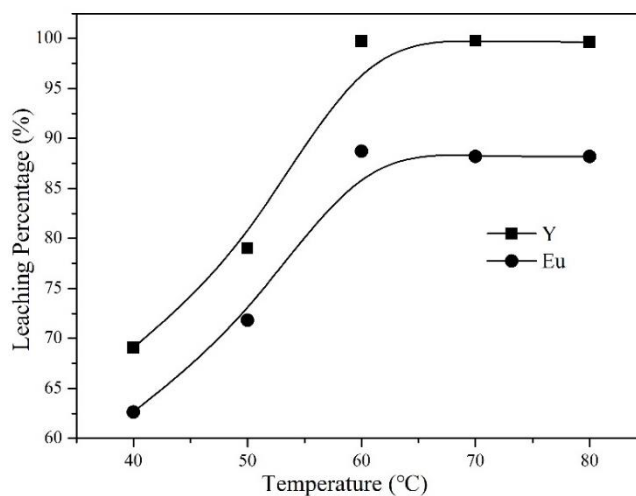

Fig. 5. Effect of temperature on leaching percentage of Y and Eu
(acid: 4 M HCl, H_2O_2: 0.2 cm³/g, t: 180 min, L/S: 10 cm³/g**)**
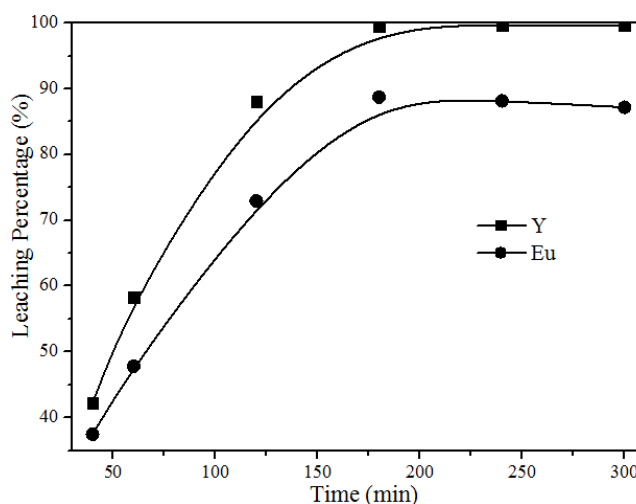

Fig. 6. Effect of reaction time on leaching percentage of Y and Eu
(acid: 4 M HCl, H_2O_2: 0.2 cm³/g, T: 60 ℃, L/S: 10 cm³/g**)**

3.5 Effect of L/S on leaching percentage of Y and Eu

The effect of L/S (2.5-12.5 cm³/g) on the leaching percentage of Y and Eu was studied using 4 M HCl and 0.2 cm³/g H_2O_2 at 60 °C for 180 min (Fig. 7). The leaching percentages of Y and Eu were increased from 19.52 to 99.48% and from 18.12 to 88.72%, respectively, with the increase in L/S from 2.5 to 7.5 cm³/g. The leaching efficiency was high at high L/S owing to the presence of a sufficient amount of acid to react with the rare earth elements. Whereas at low L/S, insufficient acid decreased the viscosity of the slurry, which resulted in the decreased mass transfer effect and low leaching percentage of the rare earth elements in the solution. Thus, at L/S of 7.5 cm³/g, the maximum leaching percentage was obtained with the recovery of 99.48% Y and 88.72% Eu. With an increase in L/S to 12.5 cm³/g, the leaching percentage of Y and Eu showed no further increase. Hence, 7.5 cm³/g was considered the optimum L/S for the experiments.

Compared with results that have already been published, the current study can reach better leaching percentages for Y (99.87%), and the addition of H_2O_2 can increase the leaching percentage of Eu significantly. The leaching percentages of Ce and Tb were respectively 0% and 0.08%, achieving the selective leaching of rare earth elements. Sulfuric acid was avoided in the present study because of the

secondary waste streams and low solubility of $Y_2(SO_4)_3$ compared with YCl_3, as well as the fact that HCl solutions can better dissolve Y compared to sulfuric acid solutions (Li., 2010).
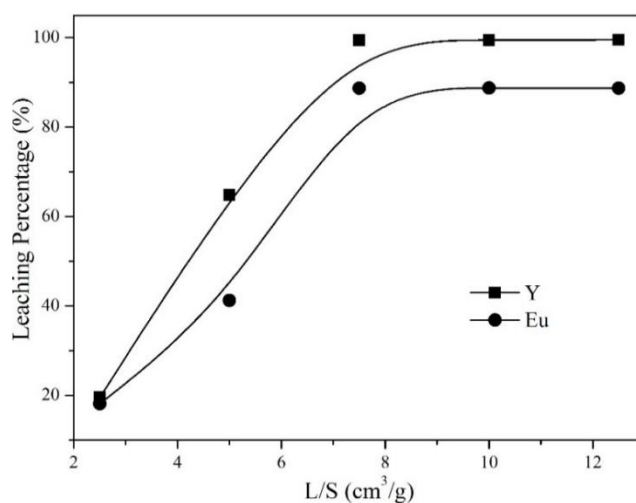

Fig. 7. Effect of L/S on leaching rate of Y and Eu (acid: 4 M HCl, H_2O_2: 0.2 cm³/g, T: 60 ℃, t: 180 min)

3.6 Kinetics of leaching

Leaching process of Y and Eu from waste phosphors using HCl solution in the presence of H_2O_2 is a typical heterogeneous reaction in liquid-solid systems: $(Y_{0.95}Eu_{0.05})_2O_3+6HCl=1.9YCl_3+0.1EuCl_3+3H_2O$. The shrinking sphere/core models as well as the Avrami model can interpret the heterogeneous reaction kinetics for most metals.

The shrinking core model considers that the rate-controlling step of the dissolution process is either the diffusion of the lixiviant through the ash layer existing on the solid particles, which could also be the diffusion through a formed solid product layer, or the surface chemical reaction on the solid particles (Levenspiel, 1972). Assuming that the particles of red phosphors have a spherical geometry and that the process is controlled by diffusion through the product layer, the integrated equation of the model can be expressed as follows:

$$1 - (2/3)x - (1 - x)^{(2/3)} = k_d t. \tag{3}$$

When the process is chemically controlled on the surface, the integrated Eq. (4) of the shrinking core model can be given as:

$$1 - (1 - x)^{(1/3)} = k_c t, \tag{4}$$

where x is the leaching percentage of Y and Eu; k_d and k_c are the rate constants for diffusion and chemical controlled processes, respectively; and t is the reaction time. According to Eqs. (3) and (4), the plots of $1-(2/3)x-(1-x)^{(2/3)}$ versus t and $1-(1-x)^{(1/3)}$ versus t would show straight lines with the slopes k_d and k_c, respectively.

To analyze the leaching kinetics of Y and Eu from waste phosphors, an experiment was conducted at different reaction times in the temperature range 40–70 ℃ (Figs. 8 and 9); the results were fitted against both models. The plots of $1-(1-x)^{(1/3)}$ versus t at different temperatures show that the kinetic data fitted well to the chemical control model (Figs. 10 and 11). This is evident from the plots (straight lines) and the high R^2 values (>0.99) obtained for Y and Eu at different temperatures. The values of the specific rate constants (k_c) obtained from the kinetic plots were used to construct an Arrhenius plot (Fig. 12). The activation energies (E_a) for the leaching of Y and Eu were calculated as 42.35 and 33.28 kJ/mol in the temperature range 40–70 °C.

Usually the surface chemically controlled leaching process has an activation energy greater than 41.8 kJ/mol. Although the activation energy of Eu was lower than 41.8 kJ/mol, the plots of the kinetic equation and the corresponding correlation coefficients (>0.98) are best represented by the chemical control model. The results obtained from this investigation strongly support that leaching of Y and Eu from waste phosphors is governed by the shrinking sphere model's surface chemical control process.
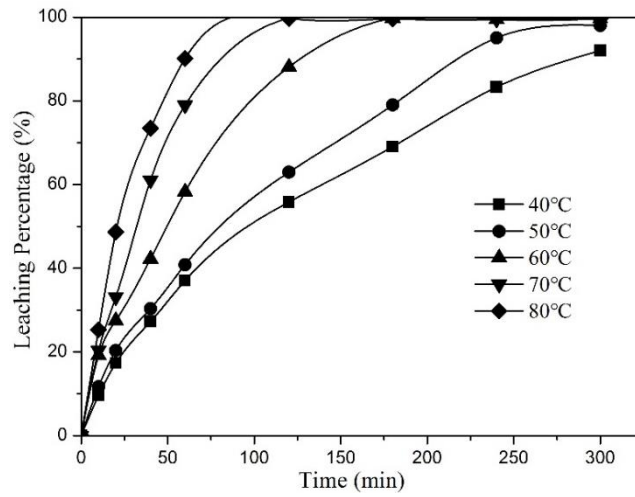
Fig. 8. Effect of temperature on leaching percentage of Y at different reaction times
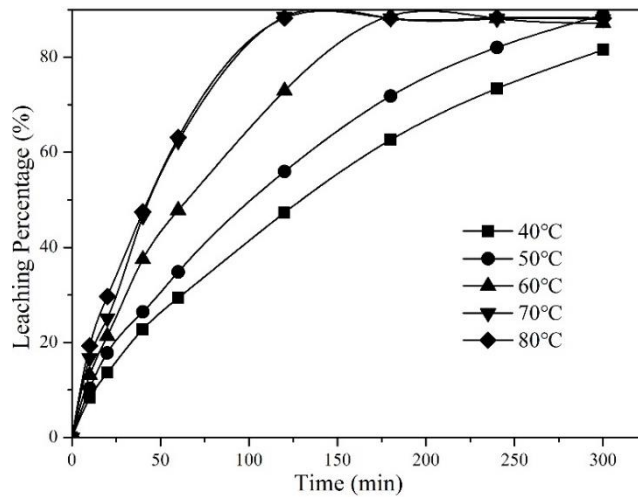(acid: 4 M HCl, H_2O_2: 0.2 cm³/g, L/S: 7.5 cm³/g)

Fig. 9. Effect of temperature on leaching percentage of Eu at different reaction times (acid: 4 M HCl, H_2O_2: 0.2
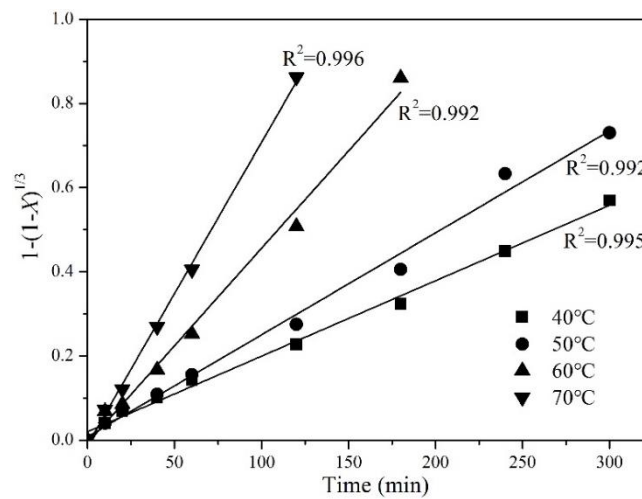cm³/g, L/S: 7.5 cm³/g)

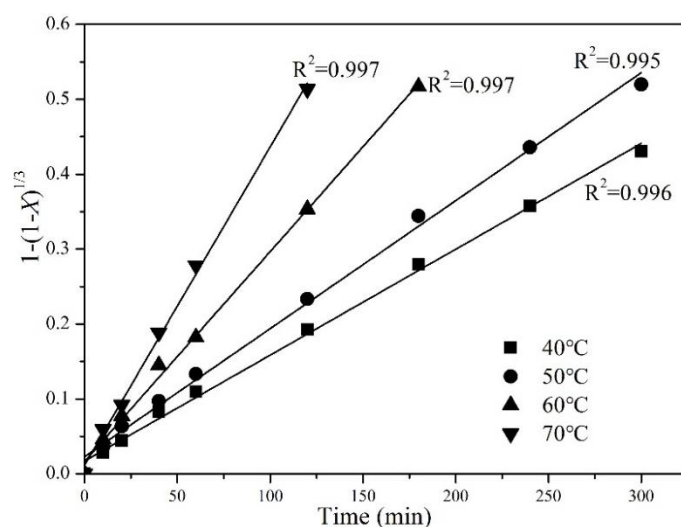Fig. 10. Chemical controlled kinetic model of leaching of Y at different temperatures

Fig. 11. Chemical controlled kinetic model of leaching of Eu at different temperatures
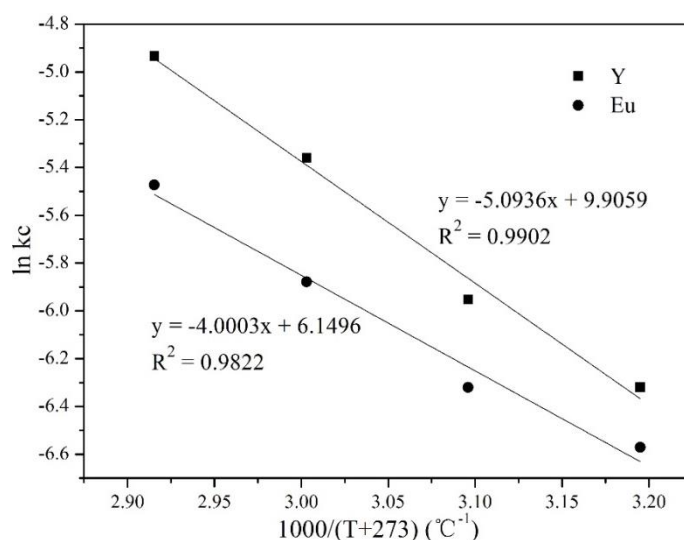

Fig. 12. Arrhenius plot for leaching of Y and Eu from waste phosphors in temperature range 40-70 ℃

3.8 Variation of phase in the leaching process

The mechanism of leaching Y and Eu from waste phosphors was established by analyzing phase changes and morphological features during the leaching process using XRD and SEM-EDS. The XRD patterns for the phase analysis of the origin waste phosphors and the residues generated during leaching are shown in Fig. 13, which exhibit gradual reduction in the peaks of $(Y_{0.95}Eu_{0.05})_2O_3$ (red phosphor). After leaching for 30 min, the major phases were $(Ce_{0.67}Tb_{0.33})MgAl_{11}O_{19}$ (green phosphor), $(Ba_{0.9}Eu_{0.1})Mg_2Al_{16}O_{27}$ (blue phosphor), and SiO_2 (quartz) and the peaks of red phosphors were significantly reduced compared with those of the untreated phosphors. After 180 min of leaching, no red phosphor phases were identified, which substantiated the dissolution of most red phosphors during the process.

The SEM-EDS analysis (Fig. 2, Table 2) of the untreated phosphors clearly shows the presence of Y and Eu elements, whereas the SEM-EDS analysis (Fig. 14) of the residues obtained at 60 ℃ under the optimum conditions shows that the main elements present in the residue are O, Si, and Al. The peaks of Y and Eu disappeared after leaching, indicating that Y and Eu elements present in the phosphors were efficiently extracted into the leaching solution.
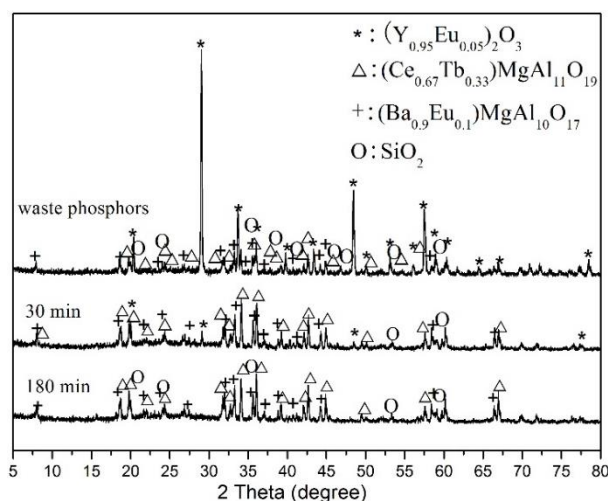
Fig. 13. XRD analysis of waste phosphors and leaching residue at 30 min and 180 min
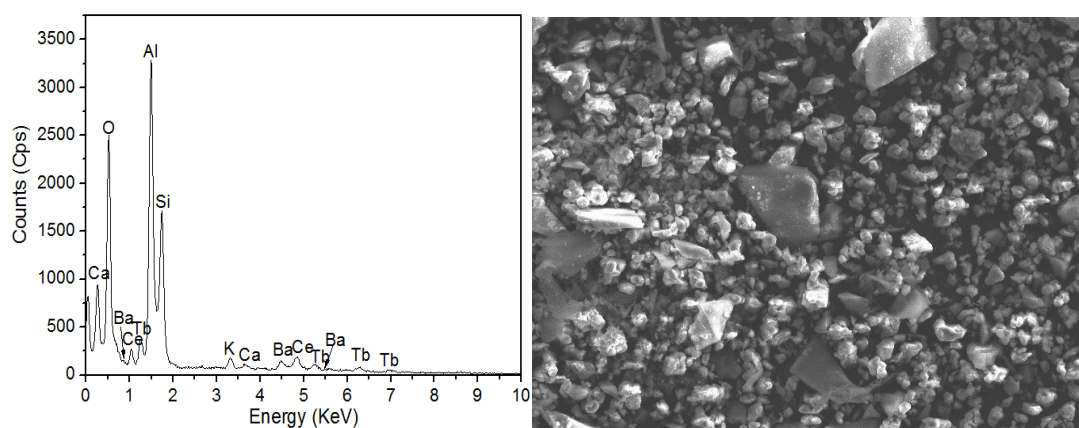

Fig. 14. SEM-EDS analysis of leaching residue at 180 min

4. Conclusions

Leaching kinetics of Y and Eu from waste phosphors was investigated. The XRD analysis of the waste phosphors indicated that the main crystalline phases of Y and Eu were Y_2O_3:Eu^{3+} (red phosphor), respectively. The optimum conditions for leaching Y and Eu from waste phosphors were determined as: 4 M HCl, 60 °C temperature, and 180 min reaction time at a 7.5 cm^3/g L/S. The maximum recoveries of Y and Eu under these conditions were found to be 99.87% and 88.72%, respectively. The leaching results obtained under different temperatures followed a shrinking sphere kinetic model: $1-(1-x)^{1/3}=k_c t$. The activation energies for leaching were found to be 42.35 and 33.28 kJ/mol for Y and Eu, respectively, in the temperature range 40-70 ℃.

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