



Chelator-induced recovery of rare earths from end-of-life fluorescent lamps with the aid of mechano-chemical energy

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ABSTRACT

Rare-earths (REs) are key components for the transition to a greener energy profile and low carbon society. The elements turn out to be of limited availability in the market, due to the supply-demand issues, exponential price rises, or geopolitics, which has led to a focus on the exploration of secondary sources for RE reclamation. End-of-life (EoL) nickel-metal hydride batteries, permanent magnets, and fluorescent lamps (FL) have been the primary sources for recyclable REs, while the recovery of REs in EoL FL (Ce, Eu, La, Tb, or Y) includes comparatively fewer processing steps than the other potential sources. In the current work, we proposed a simple, energy-efficient protocol for EoL FL processing, using chelators in combination with ball milling. The parameters for optimum chelator-assisted recovery (chelator concentration, solid-to-liquid ratio, solution pH), and milling variables (ball size, ball weight, milling speed, milling duration), were evaluated at room temperature (RT, 25 ± 2 °C). The dissolution of REs with ethylenediaminetetraacetic acid (EDTA), ethylenediaminedisuccinic acid, methylglycinediacetic acid, or 3-hydroxy-2,2'-iminodisuccinic acid, was compared at RT, while EDTA was used as the reference chelator throughout. Increasing the system temperature from 25 to 135 °C achieved at least double Eu and Y recovery, relative to that at RT, whereas the recovery rate improvement for Ce, La or Tb was insignificant. Mechano-chemical treatment at RT, via wet milling of EoL FL, with chelators, yielded a five order of magnitude increase in Ce, La and Tb recovery, however, plus a two-order increase for Eu or Y, compared with non-abetted operating conditions. It was also found that higher impact energy achieved improved recovery over a reduced milling duration with this technique having the added advantage of minimal acid consumption and reduced effluent production.

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1. Introduction

A group of seventeen elements with similar chemical and physical properties, lanthanides, yttrium, and scandium, has been designated as rare earth elements or rare earths (REs) (Migaszewski and Gałuszka, 2015). Their unique magnetic and optical properties have been advantageous for high-technology device componentry, while large volumes are continually consumed in clean energy and defense technologies (Goonan, 2011; Gosen et al., 2014). Despite

their relative abundance in the earth-crust, minable REs are less common than most other ores (Gambogi, 2018). In addition, a geographical monopolistic scenario in the distribution of exploitable RE-resources, and the regulatory, economic or political barriers to access, have led to the attachment of a 'strategically critical resource' tag to REs (Dutta et al., 2016; Fernandez, 2017; Paulick and Machacek, 2017).

The RE demand-supply gap has led to a greater focus on 'technospheric mining', which included recycling of RE scraps from the pre-consumer manufacturing process, urban mining of end-of-life (EoL) RE effluent products, and landfill mining of waste residues containing REs (Brunner, 2011; Binnemans et al., 2013; Jones et al., 2013). Such efforts might also minimize the environmental risks from the radioactive by-product thorium, which can be

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released during the mining and processing of REs (Campbell, 2014), or the so-called ‘balance problem’ in RE-production (Binnemans and Jones, 2015).

EoL nickel-metal hydride batteries, permanent magnets and fluorescent lamps (FL) have been recycled for REs so far (Gambogi, 2018), cumulatively representing more than 80% of the RE market in terms of value (Binnemans et al., 2013; Dutta et al., 2016). However, recycling REs from EoL FL is more straightforward than the other EoL resources (Binnemans et al., 2013). The EoL FL as a recyclable RE resource has either been ratified by legal frameworks (e.g., Japan), added into the long-term strategic plans (e.g., China), or defined and directed towards recovery pathways using public websites (e.g., Britain, Canada and so forth) (Wu et al., 2014).

Fluorescent lamps not only possess higher lighting efficiency, but can also save $\geq 65\%$ energy in comparison with traditional incandescent lamps, which prompted the global increase in FL use. Consequently, after an average lifespan between 7 and 24 hr, a large amount of EoL FL has been consigned to waste (Tan et al., 2015). The REs in EoL FL are contained within the inner part of a thin layer coating of trichromatic phosphors [red-phosphor, $Y_2O_3:Eu^{3+}$ (YOX); blue-phosphor, $BaMgAl_{10}O_{17}:Eu^{2+}$ (BAM); green-phosphor, $LaPO_4:Ce^{3+}, Tb^{3+}$ (LAP), $(Gd, Mg)_5B_5O_{12}:Ce^{3+}, Tb^{3+}$ (CBT), $(Ce, Tb)MgAl_{11}O_{19}$ (CAT)] (Ronda, 1997; Jüstel et al., 1998; Ippolito et al., 2017). A standard EoL FL contains about 4 g of trichromatic phosphors (Raposo et al., 2003), in the ratio: red-phosphor > green-phosphor > blue-phosphor (Wu et al., 2014).

The approaches evaluated so far for the recovery of REs from EoL FL phosphors have included hydro-, pyro-, and electro-metallurgical routes (Binnemans et al., 2013; Tan et al., 2015; Dutta et al., 2016; Jha et al., 2016; Tan et al., 2017; Innocenzi et al., 2018), with the hydrometallurgical approaches being reported as exhibiting more attractive operational characteristics, in terms of energy-consumption or cost (Tan et al., 2017). Hydrochloric acid (HCl), nitric acid (HNO_3), sulfuric acid (H_2SO_4), or sodium hydroxide (NaOH) have been used as leachant, either in tandem, or a mixture, for hydrometallurgical treatment of EoL FL for RE-recovery (Rabah, 2008; De Michelis et al., 2011; Innocenzi et al., 2013; Zhang et al., 2013; Tunsu et al., 2016; Ippolito et al., 2017). The high volume of acid or other corrosive extractant consumed has remained a challenge to overcome, in terms of impacts to the environment.

The mechanical activation of EoL FL, via treatment in a ball mill, has been proposed as a pretreatment option to the solvent-assisted leaching, to address the resistance of trichromatic phosphors to extractants (Tan et al., 2016; Song et al., 2017; Tan et al., 2017; He et al., 2018). Ball mills are simple, rapid, and economical means to provide mechanical energy in mechano-chemical processes, and can be used to modify and enhance hydrometallurgical approaches for metal recovery from waste materials (Tan and Li, 2015). The introduction of mechanical energy has been anticipated to induce lattice defects, amorphization, particle size reduction, specific surface area increase, and so forth, in crystallites (Ou et al., 2015; Tan and Li, 2015). However, the effect of mechano-chemical energy during the wet milling of EoL FL cullents using ball mills has not yet been examined.

Aminopolycarboxylate chelators, which were acclaimed for their superior extraction performance on rare metals or potentially toxic elements from solid waste (Begum et al., 2012b, 2013b; Hasegawa et al., 2013a, 2013b, 2014; Rahman et al., 2015; Begum et al., 2016; Sawai et al., 2016, 2017), were introduced as an alternative leachant in the current work, to reduce the hazard level from corrosive extractants. The chelator-assisted recovery process was aided with mechano-chemical energy, to study the effect of wet milling on RE recovery.

2. Experimental

2.1. Materials

2.1.1. Instruments

An inductively-coupled, plasma, optical emission spectrometer (ICP-OES; iCAP 6300, Thermo Fisher Scientific, Waltham, MA) was used to measure element concentration in the solutions. The ICP-OES was operated using 1.15 kW radio frequency power at the EMT duo quartz torch, 12 L min^{-1} plasma gas flow, 1 L min^{-1} auxiliary gas flow, and 0.5 L min^{-1} nebulizer gas flow, with 30 s integration duration ($n = 3$).

A high-performance, liquid chromatography (HPLC) system (TOSOH 8020; Tosoh, Tokyo, Japan) was used to quantify chelator concentration in solution. The HPLC system included a pump (DP-8020), an auto sample injector (AS-8021), a column oven (CO-8020), a UV-VIS detector (PD-8020), octadecyl silica columns (TSK-gel ODS-80TM), and data processing software (PD-8020). The mobile phase solution (ammonium dihydrogen phosphate, 5 mmol L^{-1} , pH 2.4, injection volume 20 μL) was pumped at 0.5 mL min^{-1} flow-rates (25 °C), for detection at 254 nm.

A microwave reaction system (Multiwave 3000; Anton Paar GmbH, Graz, Austria) was used for sample digestion, at a hyperbaric high temperature condition, to facilitate total element content analysis. The microprocessor-controlled reaction system (maximum microwave output power: 1400 W) had a simultaneous pressure sensor for all digestion vessels (number of vessels, 8; volume, 100 mL; vessel design, ceramic pressure jacket with PTFE-TFM liner), and the vessels were able to cope with a six MPa, controlled operating pressure, up to 260 °C for 2 h.

A Digiprep Jr block heater (SCP Science; Quebec, Canada), and an autoclave (MLS-3750; Sanyo Electric, Osaka, Japan) were used to study the effect of system temperature.

A rotary ball mill equipped with a ceramic pot mill (BP-1; AS ONE, Osaka, Japan), combined with a mill pot rotator (AV-1; Asahi Rika, Chiba, Japan) and a high-energy planetary ball mill (Mono Mill P6; Fritsch GmbH, Idar-Oberstein, Germany), were used for the mechano-chemical processing of the samples. A mini-sieve shaker (MVS-1 N; AS ONE, Osaka, Japan), a reciprocal shaker (SHK-U4; Iwaki Glass Co., Tokyo, Japan), and a filtration assembly containing a suction pump (MAS-1; AS ONE, Osaka, Japan), and cellulose membrane filters (pore size, 0.45 μm ; Advantec, Tokyo, Japan), were also used during sample processing.

A laser-scattering, particle size distribution analyzer (Partica LA-950V2; Horiba Scientific, Kyoto, Japan) was used for the measurement of particle size distribution in pulverized samples, at a wavelength of 532 nm. A water purification system (Arium Pro; Sartorius Stedim Biotech GmbH, Göttingen, Germany) was used to produce ultrapure water (resistivity, 18.2 M Ω cm^{-1}), as used for solution preparation, or dilution, or washing. A digital pH meter (Navi F-52; Horiba Instruments, Kyoto, Japan) was used for solution pH measurements.

2.1.2. Chemicals

Analytical reagent grade chemicals were used throughout, without any further purification. EDTA (ethylenediaminetetraacetic acid; Kanto Chemical, Tokyo, Japan), EDDS ([S,S]-ethylene diaminedisuccinic acid; Chelest, Osaka, Japan), MGDA (methylglycinediacetic acid; BASF, Ludwigshafen, Germany), and HIDS (3-hydroxy-2,2'-iminodisuccinic acid; Nippon Shukubai, Tokyo, Japan) were the aminopolycarboxylate chelators used for the solvent-assisted extraction of REs from EoL FL. Working solutions of HCl, HNO_3 , or NaOH (Kanto Chemical, Tokyo, Japan), as used

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