ARTICLE IN PRESS

Waste Management xxx (2016) xxx–xxx

Contents lists available at ScienceDirect

Waste Management

journal homepage: www.elsevier.com/locate/wasman

Carbon footprint assessment of recycling technologies for rare earth elements: A case study of recycling yttrium and europium from phosphor

Allen H. Hu [a,*], Chien-Hung Kuo [a], Lance H. Huang [a], Chao-Chin Su [b]

[a] *Institute of Environmental Engineering and Management, National Taipei University of Technology, Taiwan, ROC*
[b] *Department of Molecular Science & Engineering, National Taipei University of Technology, Taiwan, ROC*

ARTICLE INFO

Article history:
Received 20 July 2016
Revised 19 October 2016
Accepted 21 October 2016
Available online xxxx

Keywords:
Life cycle assessment
Carbon footprint
Rare earth recycling technologies
Phosphor

ABSTRACT

Rare earth elements are key raw materials in high-technology industries. Mining activities and manufacturing processes of such industries have caused considerable environmental impacts, such as soil erosion, vegetation destruction, and various forms of pollution. Sustaining the long-term supply of rare earth elements is difficult because of the global shortage of rare earth resources. The diminishing supply of rare earth elements has attracted considerable concern because many industrialized countries regarded such elements as important strategic resources for economic growth. This study aims to explore the carbon footprints of yttrium and europium recovery techniques from phosphor. Two extraction recovery methods, namely, acid extraction and solvent extraction, were selected for the analysis and comparison of carbon footprints. The two following functional units were used: (1) the same phosphor amounts for specific Y and Eu recovery concentrations, and (2) the same phosphor amounts for extraction. For acid extraction method, two acidic solutions (H_2SO_4 and HCl) were used at two different temperatures (60 and 90 °C). For solvent extraction method, acid leaching was performed followed by ionic liquid extraction. Carbon footprints from acid and solvent extraction methods were estimated to be 10.1 and 10.6 kg CO_2 eq, respectively. Comparison of the carbon emissions of the two extraction methods shows that the solvent extraction method has significantly higher extraction efficiency, even though acid extraction method has a lower carbon footprint. These results may be used to develop strategies for life cycle management of rare earth resources to realize sustainable usage.

1. Introduction

Rare earth elements (REEs) consist of 17 elements in the periodic table, 15 of which have atomic numbers within the range of 57–71. The REEs can be classified into light REEs (with atomic number within the range of 57–62) and heavy REEs (with atomic number within the range of 63–71). Promethium (61) is obtained by synthetic methods and does not exist in nature (Damhus et al., 2005). REEs have become among the most important strategic resources and are called the “21st century industrial vitamins” (Alonso et al., 2012). REEs are critical to high-technology applications, such as high-intensity magnets for electronic and electronic motors, efficient lighting, wind power, and hybrid electric vehicles (Hoenderdaal et al., 2013). REEs are also key ingredients in the development of environmentally friendly energy technology and economy (Tunsu et al., 2016). However, sustaining the long-term supply of these metals is difficult because of the global shortage of REE resources, which have attracted considerable concern in recent years.

According to the United States Geological Survey, approximately 99 million tons of REE reserves are available worldwide (Chen, 2011). The gross volume of global export in REEs increased from 259 t in 1990 to a peak of 111,373 t in 2004 but gradually declined until 2012. Approximately 97% of the global REE supply is produced by China, which has recently performed copious cuts of its exports to protect its environment and national downstream industries (Massari and Ruberti, 2013). The REE export quotas have been sharply restricted, and the cut export quotas was almost 70% in 2010. The total exports of REEs in 2014 reached 75,768 t, which considerably increased from 64,000 t in 2013. In 2014, China exported approximately 34,168 t of REEs, and this value was more than 51% of the total REE worldwide. The price of REEs has suddenly increased because of the reduced export quotas. The average

---

* Corresponding author.
E-mail address: allenhu@mail.ntut.edu.tw (A.H. Hu).

http://dx.doi.org/10.1016/j.wasman.2016.10.032
0956-053X/© 2016 Published by Elsevier Ltd.

Please cite this article in press as: Hu, A.H., et al. Carbon footprint assessment of recycling technologies for rare earth elements: A case study of recycling yttrium and europium from phosphor. Waste Management (2016), http://dx.doi.org/10.1016/j.wasman.2016.10.032

price of REEs in 2007 was approximately USD 13/kg, but this amount increased to USD 70/kg in 2011 (Mancheri, 2015). In 2008–2011, the prices of REO and metals increased remarkably. By comparison, their prices decreased sharply in the first half of 2012 and declined further in the second quarter of 2013 because of low demands (Fig. 1). The price of europium metal increased from $475/kg in 2008 to $2973/kg in 2011 but decreased from $2074/kg in 2012 to $954/kg in 2013 (Humphries, 2012). Financial markets could already have considered REE supply shortage into clean energy companies (Baldi et al., 2014). This fact has significantly increased REE prices, thus causing tension and high costs among high-technology industries in Taiwan (Hou, 2011). Therefore, improving the REE recycling technology in Taiwan is necessary.

Recycling techniques for REE extraction and leaching from waste products, such as phosphor, iron ore, coal fly ash, and other by-products of rare-earth materials, include acid leaching, solvent extraction, and ionic liquid (IL) methods (Gasser and Aly, 2013; Resende and Morais, 2010; Vander Hoogerstraete et al., 2013). The recovery methods of REEs via recycling of waste products, such as batteries, computer monitors, magnets, and phosphor, are shown to be promising at the laboratory scale. However, current extraction methods have caused potential toxic damage and environmental impact (Koltun and Tharumarajah, 2014). Moreover, during the life cycle of REEs, all stages consume large amounts of water, reagents, electricity, and fuel; these stages include extraction, separation, refining, and recovery from waste, specifically ion-exchange, acid leaching, and liquid–liquid extraction phases (Ecclestone, 2010). This observation illustrates that life cycle perspective plays an important role in assessing the environmental impacts REEs (McLellan et al., 2013).

Almost 22% of global electricity consumption is used for lighting, and almost 10% of REEs are made for phosphor (Roskill Information Services Ltd., 2011). The REO demand in phosphors scale will increase in the future (Rollat et al., 2016). Following the development of environmentally friendly lighting technology, the demand for phosphor, which is applied to display screens, LED, and OLED, will also increase rpidly. Therefore, developing a more efficient recycling technology for REE from phosphor is critical, especially for yttrium. The significance of Y has increased the development of technologies for recovery form virgin materials and secondary sources in recent years (Innocenzi et al., 2014). However, the current recycling technology focuses on existing technologies, which often contradict environmental protection and ignore sustainability. Thus, this technology should follow the 3R concept for actively solving technical bottlenecks and prioritize environmentally friendly technology (Schüler et al., 2011).

In light of climate change phenomenon, REE recovery from phosphor should focus on carbon footprint to reduce carbon emissions (Binnemans et al., 2013). Carbon footprint assessment can provide reference in selecting more environmentally friendly processes for REE recycling from phosphor.

In the recent years, almost all articles on rare earth recycling from phosphor have focused on the development of novel extract technologies and discussed the efficiency of rare earth recovery. However, few articles included LCA for rare earths at the rare earth recycling scale, estimated the environment effect, performed carbon footprint analysis, and provided suggestions to make recycling technologies more environmentally friendly. Most of the recent analyses have been "cradle to gate" (Koltun and Tharumarajah, 2014; Weng et al., 2016). Therefore, this paper aims to assess the carbon footprints of two recovery technologies, namely, acid extraction and solvent extraction, for recycling two REEs (yttrium

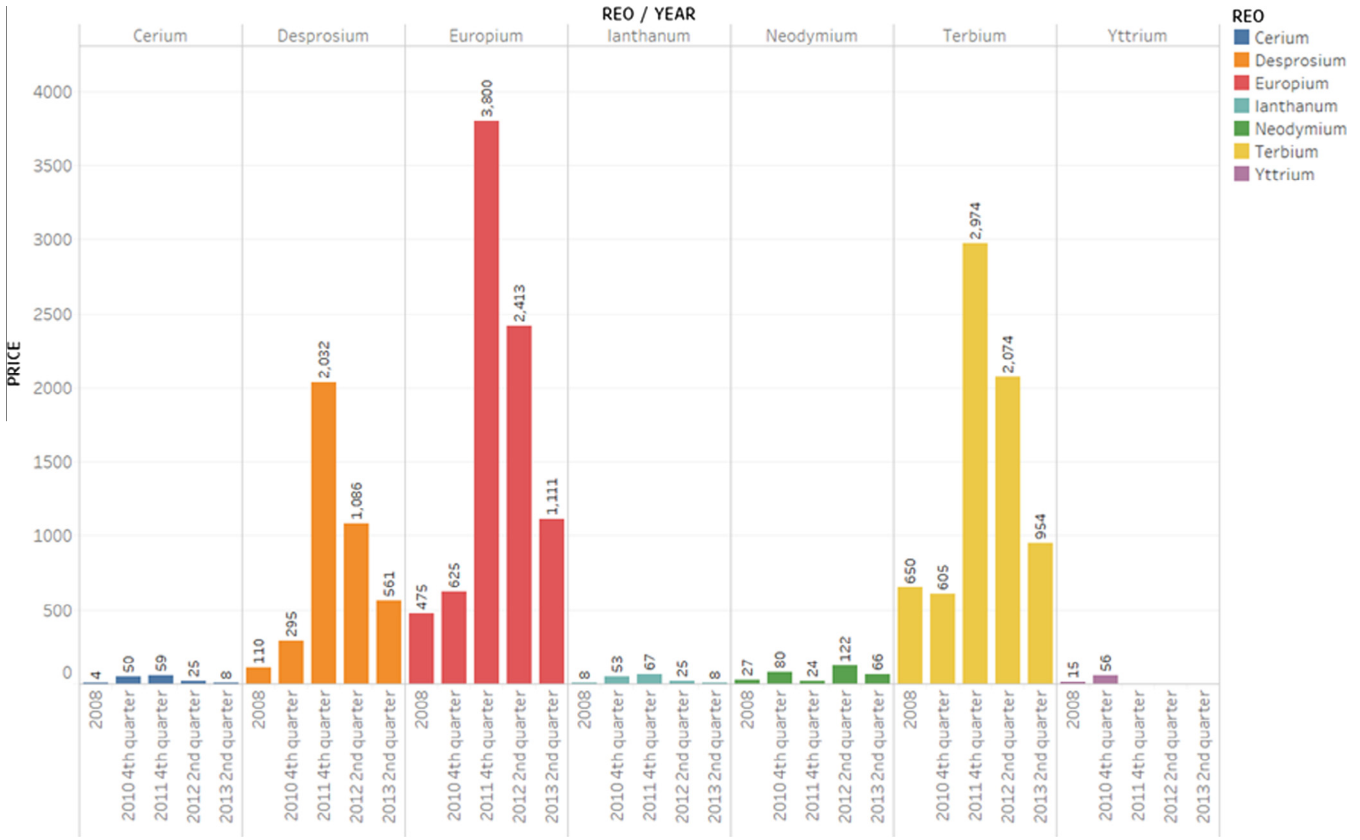

Fig. 1. Selected rare earth oxide prices, 2008–2013 (US/kg) (Humphries, 2012).

Please cite this article in press as: Hu, A.H., et al. Carbon footprint assessment of recycling technologies for rare earth elements: A case study of recycling yttrium and europium from phosphor. Waste Management (2016), http://dx.doi.org/10.1016/j.wasman.2016.10.032