Journal of Cleaner Production 51 (2013) 1–22

Contents lists available at SciVerse ScienceDirect

Journal of Cleaner Production

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

Review

Recycling of rare earths: a critical review

Koen Binnemans [a,*], Peter Tom Jones [b], Bart Blanpain [b], Tom Van Gerven [c], Yongxiang Yang [d], Allan Walton [e], Matthias Buchert [f]

[a] KU Leuven – University of Leuven, Department of Chemistry, Celestijnenlaan 200F, Box 2404, B-3001 Heverlee, Belgium
[b] KU Leuven – University of Leuven, Centre for High Temperature Processes and Sustainable Materials Management, Department of Metallurgy and Materials Engineering (MTM), Kasteelpark Arenberg 44, Box 2450, B-3001 Heverlee, Belgium
[c] KU Leuven – University of Leuven, Department of Chemical Engineering (CIT), Willem de Croylaan 46, Box 2423, B-3001 Heverlee, Belgium
[d] TU Delft, Department of Materials Science and Engineering, Mekelweg 2, 2628 CD Delft, The Netherlands
[e] University of Birmingham, School of Metallurgy and Materials, Edgbaston, Birmingham B15 2TT, UK
[f] Oeko-Institut e.V., Infrastructure & Enterprises Division, Rheinstrasse 95, D-64295 Darmstadt, Germany

ARTICLE INFO

Article history:
Received 8 September 2012
Received in revised form
25 December 2012
Accepted 26 December 2012
Available online 5 January 2013

Keywords:
Balance problem
Lanthanides
Rare earths
Recycling
Resource recovery
Urban mining

ABSTRACT

The rare-earth elements (REEs) are becoming increasingly important in the transition to a green economy, due to their essential role in permanent magnets, lamp phosphors, catalysts, rechargeable batteries etc. With China presently producing more than 90% of the global REE output and its increasingly tight export quota, the rest of the world is confronted with a REE supply risk. Mining companies are now actively seeking new exploitable REE deposits while some old mines are being reopened. Because of the absence of economical and/or operational primary deposits on their territory, many countries will have to rely on recycling of REEs from pre-consumer scrap, industrial residues and REE-containing End-of-Life products. REE recycling is also recommended in view of the so-called "balance problem". For instance, primary mining of REE ores for neodymium generates an excess of the more abundant elements, lanthanum and cerium. Therefore, recycling of neodymium can reduce the total amount of REE ores that need to be extracted. Despite a vast, mostly lab-scale research effort on REE recycling, up to 2011 less than 1% of the REEs were actually recycled. This is mainly due to inefficient collection, technological problems and, especially, a lack of incentives. A drastic improvement in the recycling of REEs is, therefore, an absolute necessity. This can only be realized by developing efficient, fully integrated recycling routes, which can take advantage of the rich REE recycling literature. This paper provides an overview of this literature, with emphasis on three main applications: permanent magnets, nickel metal hydride batteries and lamp phosphors. The state of the art in preprocessing of End-of-Life materials containing REEs and the final REE recovery is discussed in detail. Both pyrometallurgical and hydrometallurgical routes for REE separation from non-REE elements in the recycled fractions are reviewed. The relevance of Life Cycle Assessment (LCA) for REE recycling is emphasized. The review corroborates that, in addition to mitigating the supply risk, REE recycling can reduce the environmental challenges associated with REE mining and processing.

1. Introduction

The *rare earths* or *rare-earth elements* (REEs) are a group of 17 chemically similar metallic elements (15 lanthanides, plus scandium and yttrium). They are becoming increasingly important in the transition to a green, low-carbon economy. This is due to their essential role in *permanent magnets*, *lamp phosphors*, *rechargeable NiMH batteries*, *catalysts* and other applications (Tables 1 and 2). The increasing popularity of hybrid and electric cars, wind turbines and compact fluorescent lamps is causing an increase in the demand and price of REEs. In its landmark report *Critical Raw Materials for the European Union* (2010), the European Commission considers the REEs as the most critical raw materials group, with the highest supply risk (European Commission, 2010). As also acknowledged by the U.S. Department of Energy (DOE) in their medium-term criticality matrix, the five *most critical REEs* are neodymium (Nd), europium (Eu), terbium (Tb), dysprosium (Dy) and yttrium (Y) (U.S. Department of Energy, 2011) (Fig. 1). China is presently producing more than 90% of all rare earths, although this country possesses less than 40% of the proven reserves. China is not

* Corresponding author. Tel.: +32 16327446; fax: +32 16327992.
E-mail address: Koen.Binnemans@chem.kuleuven.be (K. Binnemans).

0959-6526/$ – see front matter © 2013 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.jclepro.2012.12.037

Table 1
Rare earths usage by application, in % (Curtis, 2010).[a]

Application	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Y	Other
Magnets			23.4	69.4			2	0.2	5		
Battery alloys	50	33.4	3.3	10	3.3						
Metallurgy	26	52	5.5	16.5							
Auto catalysts	5	90	2	3							
FCC	90	10									
Polishing powders	31.5	65	3.5								
Glass additives	24	66	1	3						2	4
Phosphors	8.5	11				4.9	1.8	4.6		69.2	
Ceramics	17	12	6	12						53	
Others	19	39	4	15	2		1			19	

[a] The percentages are estimated average consumption distribution by application; the actual distribution varies from manufacturer to manufacturer.

only specialized in the extraction of rare-earth oxides from the ores, but also in the downstream activities, i.e. the separation into the individual elements, the processing into rare-earth metals, and the production of rare-earth permanent magnets and lamp phosphors. Due to large and increasing domestic demands, China tightened its REE export quota from 50,145 tons in 2009 to only 31,130 tons in 2012. No sub-quota are given for the different rare earths, although since 2012 a difference is made between the light rare earths (La–Sm) and the heavy rare earths (Eu–Lu, Y). Scandium is not taken into account. These export quota may cause serious problems for REE users outside of China, and, hence, also for the development of a more sustainable, low-carbon economy. Moreover, it is anticipated that over the next 25 years the demand for

Table 2
Available present and future REE-containing streams for recycling.

Materials stream & application	REEs	Present/future contribution	Addressed in this review
1. Preconsumer production scrap and residues			
Magnet swarf and rejected magnets	***Nd, Dy,*** *Tb, Pr*	*Increasing*	*Yes*
REE containing residues arising during metal production/recycling	*All*		*No*
Postsmelter and Electric Arc Furnace residues	Ce, La, critical REEs	Future levels depend on End-of-Life presmelter recycling	
Industrial residues (phosphogypsum, red mud, etc.)	All	Relatively stable	
2. End-of-Life products containing…			
Phosphors	***Eu, Tb, Y*** *(Ce, Gd, La)*		*Yes*
Fluorescent lamps (straight/curved)	Eu, Tb, Y (Ce, Gd, La)	Relatively stable	
Compact fluorescent lamps (CFLs)	Eu, Tb, Y (Ce, Gd, La)	Increasing	
LEDs	Ce, Y	Increasing	
LCD Backlights	Eu, Tb, Y (Ce, Gd, La)	Relatively stable	
Plasma Screens	Eu, Tb, Y (Ce, Gd, La)	Relatively stable	
Cathode-ray tubes (CRTs)	Eu, Y	Sharply decreasing	
Others (speciality applications)	Also Tm		
Permanent NdFeB magnets	***Nd, Dy,*** *Tb, Pr*		*Yes*
Automobiles (small magnets as motors, switches, sensors, actuators, etc.)		Relatively stable	
Mobile phones (loud speakers, switches, microphones, etc.)		Relatively stable	