



Reclaiming rare earth elements from end-of-life products: A review of the perspectives for urban mining using hydrometallurgical unit operations



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ARTICLE INFO

Article history:

Received 11 November 2014
Received in revised form 6 May 2015
Accepted 13 June 2015
Available online 27 June 2015

Keywords:

Rare earth elements
Recycling
Urban mining
Waste processing
Hydrometallurgy

ABSTRACT

Due to their large variety of applications, their low supply and high demand, the rare earth elements (REEs) are presently viewed as being among the most critical chemical elements. Because of this, their potential recovery from end-of-life waste products has been extensively discussed both in society and in the scientific literature. This concept of recovering elements contained in end-of-life products, known as urban mining, is regarded as an important step in achieving a sustainable, circular society. This review article discusses the perspectives of reclaiming the REEs from various waste streams using hydrometallurgical methods. Three main streams are discussed in detail (phosphor-containing products, NiMH batteries and permanent magnets), touching on the state-of-the-art of material pre-treatment, leaching and separation of REEs and refining. Comparisons with the extraction of REEs from ores are drawn, bringing forth both the advantages and some of the disadvantages of urban mining.

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Abbreviations

The following abbreviations are used throughout this paper:

wt. %	percentage weight total
CCFL	cold cathode fluorescent lamp
CFL	compact fluorescent lamp
CRT	cathode ray tube
D2EHPA	di-(2-ethylhexyl)phosphoric acid
HDEHP	
e.g.	for example; such as
HDD	hard disk drive
HEHEHP	2-ethylhexyl-2-ethylhexyl-phosphonic acid
HEV	hybrid electric vehicle
LCD	liquid crystal display
LED	light emitting diode
NdFeB	neodymium–iron–boron (magnet)
NiMH	nickel metal hydride
REE	rare earth element
REM	rare earth metal
S:L	solid to liquid (ratio)
SmCo	samarium–cobalt (magnet)
TBP	tributyl phosphate
v/v	volume/volume
WEEE	waste from electrical and electronic equipment
w/v	weight/volume

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1. Introduction

The rare earth elements (REEs) are presently regarded as being among the most critical chemical elements. Their importance for the advancement of technology has been widely discussed and this is acknowledged in many recent reports (European Commission, 2011; UN Environment Programme and UN University, 2009; US Department of Energy, 2011). In 2010 the European Commission declared the REEs to have the highest supply risk among 41 different raw materials (European Commission, 2010). In 2014 the list was revised, this time comparing 54 non-energy, non-agricultural materials (European Commission, 2014). It was concluded, again, that the heavy REEs are the most critical, followed by the light REEs. This is in spite of their abundance in the Earth's crust when compared to other metals. The fact that the REEs are not that rare is well known. Light REEs such as lanthanum, cerium and neodymium are more common than lead. Even thulium and lutetium, the scarcest REEs, are one hundred times more abundant in the Earth's crust than gold (USGS, 2002).

There are several challenges associated with the virgin extraction, processing and global availability of REEs. The first one relates to their geological distribution. REE-containing minerals rarely occur in concentrated forms, making their exploitation difficult. The fact that the REEs often occur together in the same deposits raises additional challenges. In addition, they often occur together with either the uranium or thorium decay chains, making processing challenging in many countries due to radiotoxicity (Kilbourn, 1994).

The REEs have similar chemical properties, which translate into separation and purification difficulties. The most important aspect that allows for their individual separation is the lanthanide contraction –

the decrease in ionic radii alongside the lanthanide series, which has consequences for the strength of the complexes that REEs form in aqueous solution (McLennan, 1994). The chemical behaviour of sub-structures makes possible the recovery of REEs in groups containing adjacent elements. This behaviour, known as the *tetrad-effect*, was first described by Peppard et al. (1969) and was mentioned later in various other publications (Ekberg et al., 2012; Fidelis and Siekierski, 1971). Categorisation of lanthanides into four groups (lanthanum–cerium–praseodymium–neodymium, promethium–samarium–europium–gadolinium, gadolinium–terbium–dysprosium–holmium and erbium–thulium–ytterbium–lutetium) was noticed in various extraction systems when plotting the logarithm of certain physical chemical parameters vs. the corresponding atomic number. The breakpoints are explained by the stabilization energy related to the inter-electron repulsion energy of the 4f electrons. The half-filled 4f⁷ electron configuration of gadolinium is especially stabilized, leading to a clear breakpoint halfway across the series. Other breakpoints are usually located at 4f³–4f⁴ and 4f¹⁰–4f¹¹. The slow decrease in ionic radii across the lanthanide series results in increased strength of cation–anion, ion–dipole and ion–induced dipole interactions, which means that heavier REEs will create stronger complexes with extractant molecules compared to lighter REEs (Nash, 1993), allowing separation. The strength of yttrium complexes, a REE that has an ionic radius comparable to those of heavy lanthanoids (Greenwood and Earnshaw, 1998), is often comparable to the strength of heavier REE complexes. Despite these differences in chemical behaviour, complete separation of adjacent elements on a large scale is economically challenging and requires a large number of separation stages.

Production of REEs is often associated with environmental issues. Mining, leaching, pre-concentration, and the numerous separation stages needed to achieve the degrees of purity required in certain applications, sometimes as high as 99.999% (US Department of Energy, 2009), often lead to high amounts of secondary waste.

One major concern surrounding REEs is their limited availability on the market. From the 1960s until the 1980s the majority of the global production of REEs originated from Mountain Pass, California. In the 1990s, however, China began exporting cheaper REEs and the economic competition, together with environmental concerns, led to the closing of Mountain Pass in the early 2000s (Xie et al., 2014). Currently China has over 90% of the market share, although the country possesses less than half of the global deposits. The control over the amounts exported has led in the last few years to significant price fluctuations, especially for the REEs used in sustainable applications; neodymium, dysprosium, europium, yttrium and terbium. This has set in motion a search for new deposits of REEs, plans to exploit other known deposits and the re-opening of Mountain Pass in 2012.

The critical status of REEs is strongly tied to their high demand. This is becoming increasingly rapid due to the large variety of applications where they are used, some of which need very high specificity e.g. cerium oxide as polishing agent for glass; erbium for use in laser repeaters; and europium for red light emission (USGS, 2002). Most importantly, REEs are essential for future sustainable technologies. The five most critical REEs – neodymium, dysprosium, europium, yttrium and terbium (US Department of Energy, 2011) – are essential in green energy applications (Ronda et al., 1998; Schüller et al., 2011), as follows:

- neodymium and dysprosium are used in the manufacturing of permanent magnets, which are used in wind turbines and many other products, such as hard disk drives (HDDs), speakers and headphones;
- europium, yttrium and terbium are used alongside lanthanum, cerium and gadolinium in phosphors in low-energy fluorescent lamps;

- neodymium, yttrium, cerium, lanthanum and praseodymium are used in the manufacturing of nickel metal hydride (NiMH) batteries for hybrid vehicles.

Urban mining is a rather new concept that 'provides a systematic management of anthropogenic resources stocks and waste (products and buildings), in the view of long-term environmental protection, resource conservation, and economic benefits' (Cossu, 2013). Waste from Electrical and Electronic Equipment (WEEE) and other waste streams (e.g. construction and demolition waste, combustion residues, sludges, exhausted oils, previously landfilled waste, industrial residues and others) are considered urban mining targets for obtaining various materials. The urban mining of REEs from various end-of-life products and industrial waste streams started receiving more and more attention. Processing these streams for the recovery of the REEs they contain can provide another source of metals, alongside virgin mining. It is important to stress the fact that recycling cannot entirely replace mining and the production of metals from natural sources. This is mainly because of the imbalance between the demand for raw materials, the availability of recyclable streams and the existing difficulties in processing certain end-of-life products for the recovery of REEs. Urban mining, however, can prevent to some extent excessive mining by providing another source in addition to geological deposits. This may also reduce some of the disadvantages with regard to the time it takes to discover and exploit new/already existing deposits.

The scope of this paper is to address some of the current efforts directed towards reclaiming the REEs contained in end-of-life products. As examples of this rather wide field, some specific waste flows were selected; products containing REE-based phosphors such as fluorescent lamps and computer and television screens, products containing permanent magnets and NiMH batteries. Various scenarios for urban mining via hydrometallurgical processing are reviewed, together with state-of-the-art examples. The main advantages and disadvantages of a hydrometallurgical approach are presented, as well as future perspectives such as ionic liquid separation.

2. Process options and unit operations

2.1. Pyrometallurgical and hydrometallurgical treatment

The recovery of REEs from various streams can be carried out using hydro-, pyro- or electro-chemical processes (Meyer and Bras, 2011). The nature of the stream to be processed, together with its chemical complexity and REEs content, determines the possible treatment methods. While pyrometallurgy is widely used in the processing of high-grade ores, this method is not always suitable for low-grade ores due to its high-energy requirement, the difficulty of treating small amounts of material and additional burdens on environmental treatment expense (Yoon et al., 2014b). Moreover, most often the products that are obtained pyrometallurgically require additional processing to produce pure rare earth metals (REMs) and REE compounds. This is because gas–solid pyro-processes of streams containing more than one REE will lead to mixed products; the same being true for electroslag refining. To obtain individual REMs, the end product needs to be further processed, e.g. using hydrometallurgy or molten salt electrolysis. For REE-based end-of-life products that end up in electric arc furnaces or non-ferrous smelters, the REEs will typically be found in the slag phases in low concentration, making their economic recovery more difficult (Binnemans et al., 2013). There are advantages associated with electro-chemical methods, such as those using molten salts as a reaction medium over water. These advantages include high chemical stability, high conductivity, high reaction rates, broad applicable temperature range and low vapour pressure (Tanaka et al., 2013).

Hydrometallurgical processes have distinct advantages in separating REEs, especially when it comes to the recovery of individual elements. This applies both to the extraction of REEs from mineral ores and to

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