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A REVIEW OF A CASE STUDY ON RARE EARTH ELEMENTS RECOVERY FROM SECONDARY RESOURCES IN EUROPEAN HYDROWEEE PROJECT

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ABSTRACT

Rare earth elements (REE) are high-value raw materials and of strategic importance in various applications, including emerging technologies globally. The fast-evolving and rapid technological changes have caused the demand for REE continuously increase. In the future, conventional deposits may no longer satisfy industrial demand. Therefore, a holistic study on REE recovery from potential secondary sources must be carried out. This paper presents the recent findings on the recovery of REE from waste electrical and electronic equipment (WEEE) or e-waste as an alternative to extraction from mines. The HydroWEEE project in Italy, funded by the European Union (EU) from 2009 until 2016, is used as the case study. The future perspectives regarding the circular economy, benefits, and challenges in implementing the recovery of REE from WEEE are also discussed.

Keywords: Rare earth elements, recovery, waste electrical and electronic equipment, carbon reduction

INTRODUCTION

Rare earth elements (REE) are a set of 17 chemical elements which consist of yttrium (Y) and scandium (Sc), as well as the 15 lanthanide elements: lanthanum (La), cerium (Ce), praseodymium (Pr), neodymium (Nd), promethium (Pm), samarium (Sm), europium (Eu), gadolinium (Gd), terbium (Tb), dysprosium (Dy), holmium (Ho), erbium (Er), thulium (Tm), ytterbium (Yb), and lutetium (Lu). They are of growing interest, and their applications cover many fields such as high-tech components, green technologies and material industries of high-temperature superconductors, secondary batteries, and electric or hybrid cars.

REE are grouped depending on the atomic number, into light rare earth elements (LREE) – La, Ce, Pr, and Nd, and into ‘middle and heavy’ HREE – Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb and Lu. Minerals containing REE are differentiated into various groups depending on the content of REE: monazite, xenotime, and bastnaesite are the three most frequently extracted rare earth (RE) minerals. In addition, the interest in ion adsorption clay deposit (IAC) has gained of late as it is becoming a significant source of HREE (Chen et al. 2019).

Modern society faces the problem of a continuous increase in demand for raw materials, including mineral resources, due to the rapid population and economic growth (Nasrollahi et al., 2020). REE are included in the list of 'critical' raw materials in the United States of America (USA), the European Union (EU), and Japan. REE are considered critical because they are at risk of diminishing supply and their impact on the economy is higher than most other raw materials.

Over 90% of the world's economically recoverable REE are present in primary mineral deposits, i.e., in bastnaesite ores, which are in China (Bayan Obo) (Habib & Wenzel, 2014) and the USA (Mountain Pass in California). The monazite deposits are the second largest REE-bearing mineral and are primarily located in Australia, South Africa (Jun et al., 2010), Brazil, Malaysia (Kołodyńska &

Hubicki, 2012), India (Maitra et al., 2009) and Russia (Kalinnikov et al., 2010). China has almost half of the world's known reserves and dominates 94% of REE global production (Schlinkert & Van Den Boogaart, 2015).

The stages of REE production consist of mining, separating, refining, alloying, and manufacturing RE into end-use items and components (Borowik et al., 2012). Because of the growing demand and the desire to have no supply constraints, many countries, including Malaysia, are evaluating the potential to exploit their resources. Due to the low concentrations in the earth's crust, REE recovery from secondary resources could become a new source of supply and has been rapidly growing globally. One of the possible ways is to recover REE from Waste Electrical and Electronic Equipment (WEEE).

WEEE, also known as e-waste, is defined as any broken, non-working, old or obsolete electric or electronic appliance such as a TV, computer, air conditioner, washing machine or refrigerator. It is generally grouped into two main types, either e-waste generated from the industrial sector or household. WEEE is becoming a global issue, where the more electrical and electronic equipment is produced, the more e-waste needs to be disposed of or managed properly. In addition to containing toxic and hazardous materials such as lead, cadmium, and arsenic, WEEE also contains economically significant levels of precious and critical metals, for example, gold, copper, silver and REE. Therefore, WEEE represents a potential secondary source of valuable material, whose recovery is a growing business activity worldwide (Europian Commission, 2019).

This paper aims to review a successful case study of urban mining dealing with two research projects funded within the European FP7 framework. The HydroWEEE Project developed an innovative hydrometallurgical process to recover REE from WEEE. Furthermore, the economically convenient from a circular economy perspective and the benefits and challenges in implementing the concept of recycling and recovery REE are discussed. The assessment will enable knowledge expansion in current research to better understand the challenges in REE recovery from secondary resources for the upcoming research strategy.

CASE STUDY

In Europe, REE are categorized under the critical raw material (CRM). These raw materials are economically and strategically important for the European economy but have a high-risk associated with their supply. The REE gained international attention in 2011 when prices rose after the Chinese export quota restriction. Aside from mining, one of the alternative options for extracting REE is to recover them from waste. Therefore, further advancement in alternative processing techniques was required for the circular economy and sustainability of the REE industry. It is generally aimed to achieve economic development while respecting resource limitations.

In particular, the European Union (EU) was aimed at WEEE recycling to reduce the disposal of waste and “to contribute to the efficient use of resources and the retrieval of valuable secondary raw materials”. WEEE is the fastest-growing waste stream in Europe. These wastes represent an important secondary source of REE in Europe. Examples of electronic waste that has an important percentage of REE are spent fluorescent lamps and cathode ray tubes (CRTs). However, these devices are classified as hazardous materials for the high concentration of heavy and toxic metals and must be appropriately disposed of.

Therefore, the EU had established the directives for their disposal and recycling, given their high concentration of REE. The ideas and insights on minimizing waste and conserving essential resources inspired Relight Ltd to participate in a groundbreaking initiative funded by the European Framework Programme (FP7), called HydroWEEE (2009-2012). This project was followed by HydroWEEE-Demo (2012-2016). The outcomes of both projects were documented in the HydroWEEE Project Consortium and the HydroWEEE-Demo Project Consortium reports for the Framework Programme.

Relight Ltd was established in 1999 by a project with Philips to create a network to collect and recover fluorescent lamps in Italy. Relight has been a pioneer in collecting, treating, and recovering WEEE since 2005. It is a leading Italian company that recycles WEEE in general. The core of its business is the treatment of WEEE belonging to televisions and monitors, which accounts for 80% of the input waste, and the treatment of fluorescent lamps, which accounts for 4% of the input waste of about 824 tonnes/year. In 2014, Relight treated 600 tonnes of lamps, accounting for 54% of the annual quantity collected in Italy, and produced 15 tonnes of fluorescent powder from lamps. In addition, it has been involved in a lot of research and development regarding waste management, one of which was the HydroWEEE projects, for recovering REE, namely, Eu, Tb and Y from spent lamps and CRTs by hydrometallurgical processes and the development of HydroWEEE demo plants.

HydroWEEE Projects

The first phase of the HydroWEEE Project (2009-2012) dealt with recovering rare and precious metals from WEEE, including lamps, and spent batteries, by the hydrometallurgical process. The idea was to develop a mobile demo plant using hydrometallurgical processes to extract metals like Y, indium, lithium, cobalt, zinc, copper, gold, silver, nickel, lead, and tin in high purity (above 95%). For the content of REE, the lamps have different phosphors: the red phosphors (Y_2O_3:Eu^{3+}), the green phosphors ($LaPO_4$:Ce^{3+}, Tb^{3+}) and the blue phosphors ($BaMgAl_{10}O_{17}$:Eu^{2+}) (Jastel et al., 2008; Yu & Chen, 1995; Ronda, 1995). Figure 1 demonstrates the flow of hydrometallurgical processes developed for recycling and recovering REE from fluorescent lamps and CRTs (Innocenzi et al., 2016).

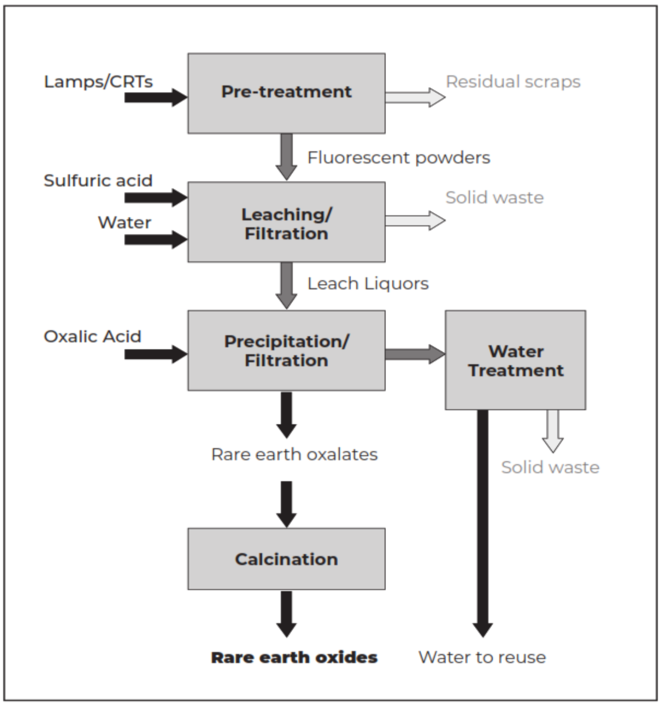

Figure 1. Hydrometallurgical process for Y and Eu recovery from lamps and CRTs

After collecting the spent lamps and CRTs, the recycling processes start with a mechanical pre-treatment of the waste, including crushing, diamond cutting technology (for CRTs) and sieving. The analysis showed that the fluorescent powders consists of Y, Eu and Tb with the concentration of 15%, 0.6% and 0.5%, respectively. For CRTs, the analysis showed that Y and Eu were, on average, 14% and 0.9%, respectively.

A mobile demo plant as shown in (Fig. 2) was developed when the hydrometallurgical processes were attained (Kopáček, 2013). The innovative process was designed as the universal process; thus, several fractions such as lamps, CRTs, LCDs, printed circuit boards and Li-batteries can be treated in the same mobile plants in batches. The development of the mobile demo plant has benefited small and medium enterprises (SMEs) as several companies can use it at different times. Therefore, the necessary quantities of waste, as well as investments, can be limited.

Figure 2. View from (right) back, (left) front side of HydroWEEE mobile demo plant

A stationary demo plant as shown in Figure 3 was set up during the second phase of the HydroWEEE Project (2012-2016) (Kopáček, 2013). The demo plant can recover REE from various electronic wastes such as batteries, LCD screens and circuit boards. However, it is most suited for the recovery of Y, Eu, Tb and other REE from fluorescent powders resulting from the exhausted lamps and CRTs recovery. The plant operated two batches per day with the capacity of producing 184.8 tonnes per year of fluorescent powder from spent lamps. The annual mass balance of the hydrometallurgical process (precipitation of REEs) is 59.7 tonnes of REOs mixture per year, consisting of 91.3% yttrium oxide (Y_2O_3), 4.07% europium oxide (Eu_2O_3), 1.08% gadolinium oxide (Gd_2O_3), 0.28% terbium oxide (Tb_2O_3), 0.11% cerium oxide (Ce_2O_3) and 0.01% lanthanum oxide (La_2O_3) (Favot & Massarutto, 2019). Innocenzi et al. (2016) reported that 93% of Y and Eu from lamps and 74% of Y from CRTs could be recovered from the process.

Figure 3. HydroWEEE Stationary Demo

CIRCULAR ECONOMY

The conventional REE processing approach to producing lamp phosphors from primary sources using conventional hydrometallurgical approaches is in the linear economy of "extract, make, consume and dispose of", as demonstrated in Figure 4 (European Commission, 2019). However, the entire process is often associated with environmental impacts and elevated human health risks due to the existence of radioactive material, thorium, and uranium, in the host material.

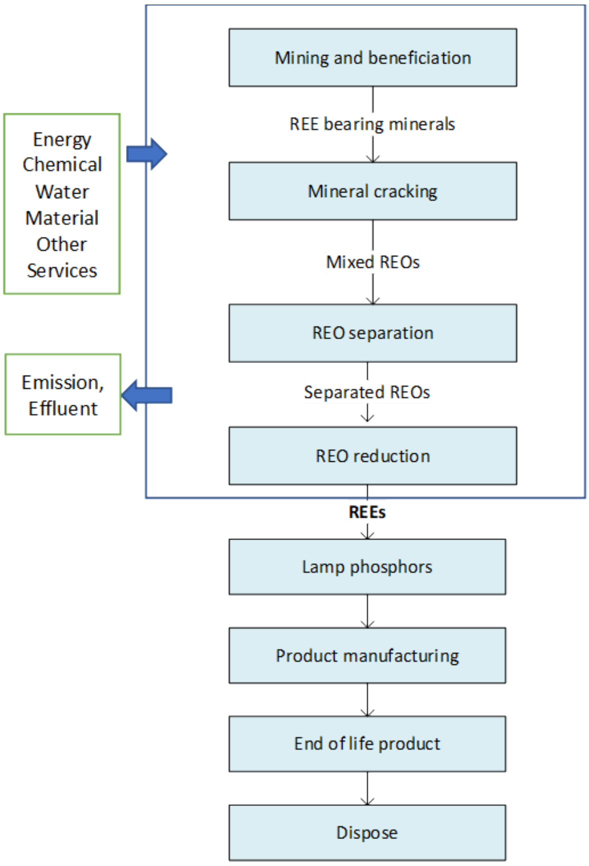

Figure 4. Linear economy in REE industry

With a revolution such as the hydrometallurgical recovery process from the HydroWEEE project, a linear economy in REE industry has transformed into a circular economy framework. This transformation has expanded the benefits of improving resource efficiency and the contribution of the circular economy to the sustainability of REE consumption. The process flow for the life cycle of REE towards the circular economy in fluorescent lamp production is shown in Figure 5 (Binnemans et al., 2013).

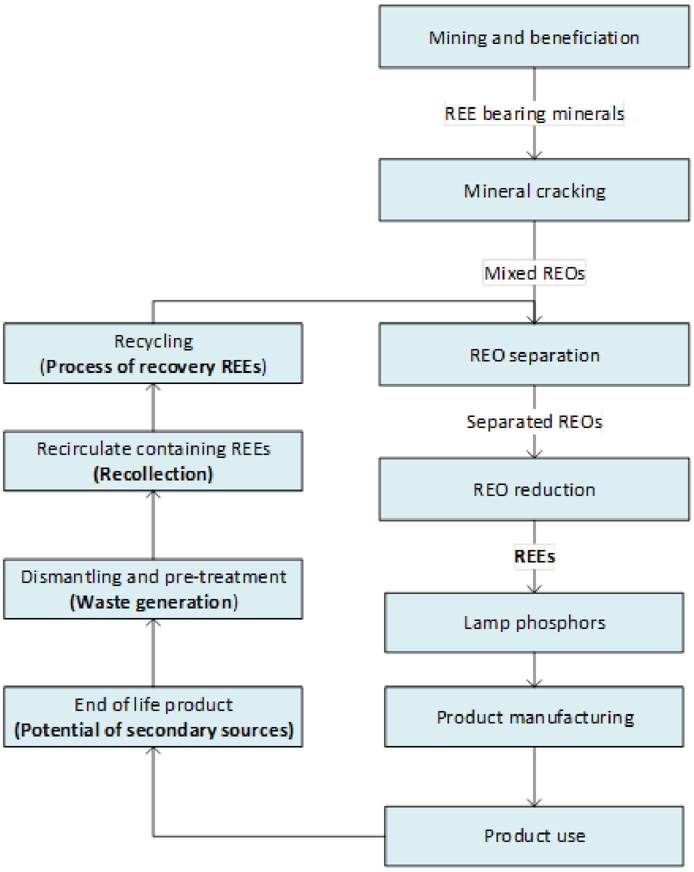

Figure 5. Recycling prospect for REEs and opportunities for sustainable development

BENEFITS OF REE RECYCLING AND RECOVERY

Recovery of the REE is the most drastic recycling method, but it delivers the purest end products in the form of high-purity rare earth oxide (REO). In this approach, the lamp phosphor fraction is considered a high-value rare-earth ore, with a high concentration of REE such as europium, terbium, and yttrium considered precious material. Therefore, they can be used for making new lamp phosphors or for making new REE-containing compounds for other applications; for example, europium can be used in security markers, terbium in magnets and yttrium in phosphors as well as in high-tech ceramics. Therefore, properly managing the REE recovery and recycling process from WEEE will benefit the environment, economy, and society.

Primarily, it may abate environmental and health problems associated with hazardous substances. Owing to the increased demand for REE, adverse environmental impacts associated with the production of REE have become a global concern. In general, REE recycling has significant advantages over the mining of rare earth, including savings in energy, water, and chemicals consumption, along with a significant reduction of carbon emissions, effluents and solid waste generation resulting from the extraction and processing of rare earth ores. REE recyclate does not contain radioactive thorium and uranium, unlike the primary mined rare-earth ores. Therefore, radioactive tailing stockpiles and mining health problems can be, at least partially, avoided. There are also possible benefits from avoiding land allocation for the mine and radioactive waste streams and transportation. Energy use (which is associated by CO_2 emissions and other greenhouse gas (GHG) emissions) and water use are typically much lower for secondary than primary material. The recycling process may reduce the environmental burdens associated with the consumption of primary new materials (Cucchiella et al., 2015).

Furthermore, the process may deliver scarce and valuable materials for the economy as the current demand for REE has significantly increased, which has caused an increase in their prices. The balance between the demand by the market and the natural abundance of the rare earth in ores is a significant problem for manufacturers of these elements. Therefore, recycling helps address the so-called “balance problem”, namely that certain REEs with high demand levels, for instance, europium, are present in small quantities in REE ores along with other REEs with low demand, such as lanthanum and cerium. In order to meet the demand for the former, the latter is produced in excess and is stockpiled. The ideal situation is a perfect match between the demand and production of rare earth so that there are no surpluses of any REE. This situation would result in the lowest market price for rare earth because all the elements share the production costs. Therefore, recycling these REEs from end-of-life fluorescent lamps can help keep the rare-earth markets balanced.

Finally, recycling may provide ancillary social benefits such as social inclusion opportunities in different ways: employment for disabled people or the long-term unemployed, helping to bridge the digital divide, and other related benefits (Kissling et al., 2012). There is likely to be less social resistance as efforts toward a circular economy for REE develop alongside their green economic uses in products.

CHALLENGES

Firstly, the ability to achieve higher recycling rates of REE from the WEEE will be disturbed due to non-systematic e-waste collection, lengthy product life span and high cost in producing the purest end-product. The WEEE represents the fastest-growing waste stream in the EU, generating about 12 million tonnes in 2019 (Forti et al., 2020). In many cases, and sometimes despite legislation, the WEEE is not collected separately for recycling but is disposed of with mixed Municipal Solid Waste (MSW). One of the limiting factors that affect the volume and availability of material for recycling is the waste leakage into the 'grey market' of informal or illegal low-technology recyclers. This problem typically leads to losing the products' small quantities of hard-to- extract metals.

Secondly, although recycling is a promising option for mitigating REEs supply issues and reducing overall environmental burdens associated with the production and consumption of these metals, it is not a short-term solutionbecause many emerging technologies that rely on REE, such as wind turbines, electric vehicles, have a long-life span and are not yet ready to be recycled. This matter will still cause a shortage of material supply as well.

Finally, the non-eco-friendly treatment and recovery processes with higher operating costs required to purify the mixtures obtained from consumer devices. For example, according to Wang et al. (2011), the REE content in the phosphors of lamps can reach 27.9%, but only 10% is currently recovered. The low percentage of REE recovery is due to the high acid resistance of some types of phosphors during the conventional recovery process that uses acid for leaching. While Favot and Massarutto

(2019) studied the economics of yttrium recycling from spent lamps, they reported that recycling is a valid option in economic terms if the market price is above 14 €/kg. Considering the external costs of mining REE, which have been computed at 4.46 €/kg, the recycling option is convenient when the price of yttrium oxide is above 9.54 €/kg. They determined that in 2012 and 2013, it was convenient to recover yttrium because its price was higher than the costs of separated waste collection plus the cost of treatment and recovery. However, between 2014 and 2016, the market price of yttrium did not cover such costs, thus making the treatment and recovery of yttrium less convenient than disposal. Therefore, the low-priced green technologies for separations systems are expected to effectively contribute to recycling REE from end-of-life rare-earth-containing products.

CONCLUSION

The detailed process analysis in the laboratory and the practical experience gained from the HydroWEEE projects will give useful indications about this new approach to the WEEE recycling business in Malaysia. In the medium to long term, the supply of REE is expected to exceed demand, except for a few essential REE such as Y, Dy, Nd, Eu, and Tb which are used in the production of high-tech consumer products, such as cellular telephones, computer hard drives, electric and hybrid vehicles. The increase in the price of REE since 2009 has increased interest in the substitution and recycling of REE. To this point, the recovery of REE from spent fluorescent lamps is the most mature in the industrial-scale application. The economics of the REE recycling determined by the costs of the process and the need to achieve economies of scale and the REE market pricing as well as the absence of a supply chain structure geared towards the pre-processing of WEEE with an emphasis on REE. Therefore, an effective supply chain for recovery and recycling is necessary to ensure the availability of suitable recyclates with high REE concentrations, thus contributing to the sustainable development of REE and the circular economy.

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