Recycling and substitution of light rare earth elements, cerium, lanthanum, neodymium, and praseodymium from end-of-life applications - A review

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Highlights
- An overview of light rare earth elements is given
- Recycling of light rare earth elements from selected end-of-life application is given
- Criticality and environmental concerns of light rare earth elements are stated
- Substitution of light rare earth elements in magnets, batteries, phosphors and catalysts is analyzed
- Sustainability benefits of light rare earth elements recycling and substitution strategies are proposed

Abstract
The light rare earth elements (LREEs) lanthanum, cerium, neodymium and praseodymium are increasingly used in renewable energy technology and are applicable in portable electronic devices, such as phosphors in lightning applications and in catalysis. The extraction of REEs from virgin ores causes environmental degradation. LREEs are considered as critical metals. To overcome the environmental and criticality challenges of LREEs, recycling presents means by which they can be obtained from secondary sources. Presently, the recycling rate of LREEs is still very low. Substitutes of LREEs in most cases are either inferior or still undiscovered. This study investigates the criticality challenges and environmental impacts of producing LREEs from virgin ores. It focuses on LREEs obtainable in selected end-of-life products considered to have significant recycling potential; these include NdFeB magnets, Ni-MH batteries, phosphors in lighting and catalysts. Current recycling technologies, including representative methods and current recycling challenges are also reviewed. Although current recycling technologies have recorded growth, there is still a need for further improvements. The article highlights current LREEs substitution advances and the faced challenges in finding suitable LREEs substitutes. Furthermore, future ways to promote sustainability of LREEs recycling, to improve substitution, and to tackle the criticality challenges of LREEs are proposed.

Keywords: Light rare earth elements, recycling, substitution, criticality, magnet, cerium

1. Introduction

Historically, rare earth elements (REEs) were first studied, documented and named "rare earth" in 1794 by Juhan Gadolin, a Finnish Chemist at Åbo Akademi University (Turku) in Finland (Haskin and Frey, 1966). The modern research on REEs was pioneered by Schmitt and his colleagues in 1960 (Schmitt et al., 1960). REEs also known as rare earth metals or simply as rare earths, are

defined as the 15 lanthanides together with scandium and yttrium (Fig. 1) (IUPAC, 2005). The title rare earth is actually a misnomer because REEs are abundant in nature and can be found in high concentrations in many minerals and in low concentrations in every part of the earth crust. (Haskin and Frey, 1966). The main reason for the name (rare) is simply that they are chemically similar to each other, occur together in nature and possess similar trivalent oxidation state (Pohl, 2011). REEs are also very difficult and costly to separate individually from its ore (Dutta et al., 2016). Additionally, REEs are rarely concentrated in minable ore deposits (Izatt et al., 2014).

The International Union of Pure and Applied Chemistry (IUPAC) has classified REEs into light rare earth elements (LREEs) and heavy rare earth elements (HREEs). This classification can be based either on their atomic weight or on their position on the Periodic Table of elements (International Union of Pure and Applied Chemistry, 2016). LREEs consist of lanthanum, cerium, praseodymium, neodymium, promethium, and samarium with the atomic numbers from 57 to 62. HREEs consist of europium, gadolinium, terbium, dysprosium, holmium, erbium, thulium, ytterbium, and lutetium with the atomic numbers from 63 to 71. (IUPAC, 2005) LREEs have a higher concentration than HREEs in the environment, which means that LREEs are more abundant than HREEs (Brumme and Brumme, 2014). REEs are soft silver metals having high melting points and they discolor rapidly when in contact with air. In nature, LREEs occur mostly in their +3-valence state, which means that they form oxides. However, cerium in an oxidizing environment forms Ce^{4+} (Gunn, 2013). LREEs possess unique and important magnetic and spectroscopic properties as well as optical features and exceptional physicochemical properties (Gupta and Krishnamurthy, 2005).

Fig. 1. IUPAC periodic table of elements. Non-lanthanide rare earth elements (REES) scandium and yttrium, light rare earth elements (LREEs), and heavy rare earth elements (HREEs) are ringed with different colours (International Union of Pure and Applied Chemistry, 2016).

Currently, there are 120 million tons (Fig. 2.) of REEs reserves worldwide (Gambogi, 2016). The total crustal abundance of REEs is estimated to be 9.2 ppm (British Geological Survey, 2011). With regards to the mine production (Fig. 3), China is the largest producer of REEs at 105,000 tons with a mine production rate of 83%, Australia is the second largest producer with 11% mine production rate. Other countries with economic amounts of REEs include Russia, Brazil, and India. (Gambogi, 2016)
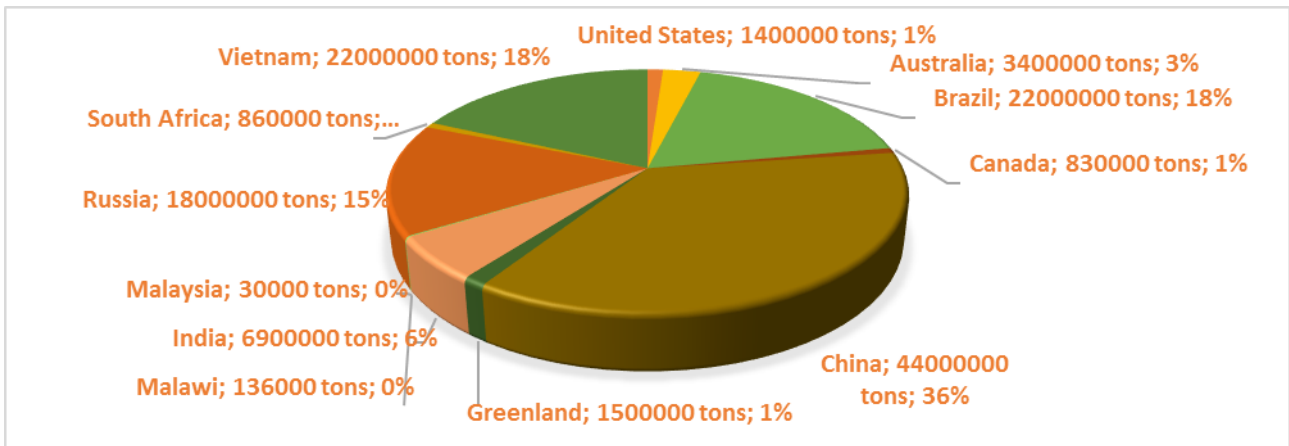
Fig. 2. Global reserves of REEs (modified from Gambogi, 2016).
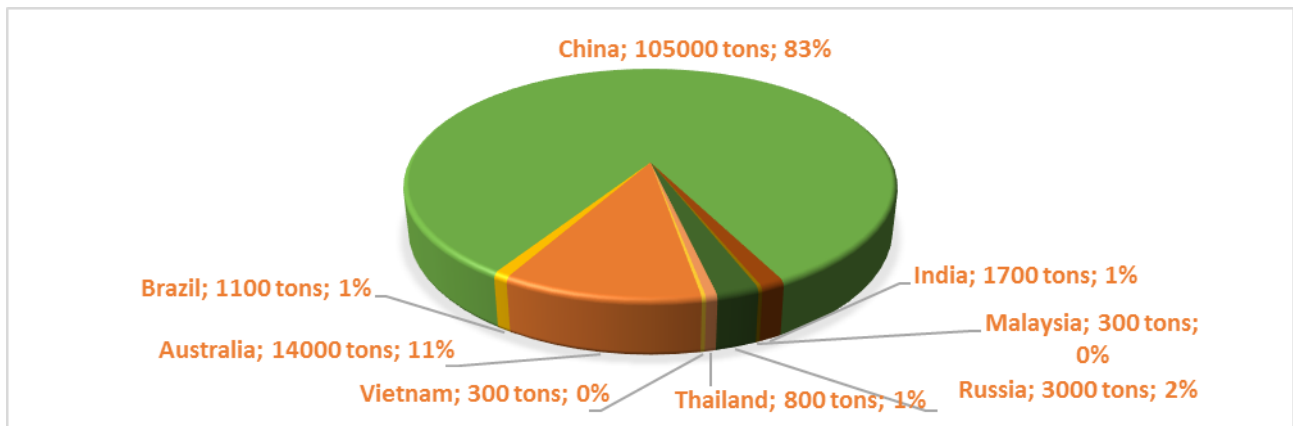
Fig. 3. Global mine production of REEs (modified from Gambogi, 2016).

Economically, the predominant sources of LREE minerals are bastnaesite, monazite, xenotime, fergusonite, loparite, apatite, and kaolinite (Sarapää et al., 2013). Bastnaesite deposits hold the biggest concentration of LREEs worldwide and are the primary source of LREEs cerium, lanthanum, neodymium, and praseodymium located in Bayan Obo REE-Nb-Fe deposit located in China (Gupta and Krishnamurthy, 2005; Wang et al., 1994). This study is mainly focusing on cerium, lanthanum, neodymium and praseodymium. The approach used is to address the questions of criticality and environmental impacts encountered when producing the LREEs from virgin ores. It also addresses the major challenges encountered when obtaining the LREEs through recycling technologies in selected applications. The end-of-life products considered have significant recycling potential; they include NdFeB magnets, Ni-MH batteries, phosphors in lighting and catalysts. In addition, current LREEs substitution advances are highlighted and challenges are investigated. Future ways to promote sustainability of LREEs recycling and ways to improve their substitution and to tackle the criticality challenges are proposed.

1.1. Selection and classification of research methods

This study is based on a compilation of several relevant peer-reviewed articles, books, conference papers, as well as government and company websites worldwide. A string of keywords such as "rare earth elements", "environmental impacts of rare earths production", "criticality of rare earth elements", "critical metals", "rare earths application", "recycling of rare earths permanent magnets", "substitution of rare earth permanent magnets", "rare earths recycling techniques", " substitution of rare earths in lamp phosphors", "recycling of rare earths", "rare earths recycling techniques", "recycling of rare earths in catalyst", " recycling of rare earths in phosphors", " recycling of rare earths in Ni-MH batteries", "substitution of rare earths in catalysts" etc, was used to search articles and texts used in the study. Some officially published literature was obtained from the US Department of Energy, European Commission, and US geological survey for mineral commodity summaries and British geological survey. This study is divided into five sections. The introductory section gives a brief history of LREEs, their classification and their global distribution. Section 2 analyses the application, criticality, and environmental impacts of LREEs mining and processing. In this section, the applications and properties of LREEs are clearly indicated. In addition, the meaning of critical metals and LREEs as critical metals are illuminated. Furthermore, the environmental impacts of LREEs mining and processing are covered. In section 3, the main recycling steps and techniques of LREEs are summarised and analysed, and their case study in selected end-of-life LREEs bearing products was considered. A filtered literature work for the selected technical process is performed. Therefore, from the ideas and experimental results in the literature, representative examples of each recycling technology are selected, summarized and reviewed in details. Recycling techniques include metallurgical techniques namely hydrometallurgical and pyrometallurgical. In section 4, what it means to have successful substitution is clearly indicated. Focus is also placed on ways substitution of LREEs can take place. In addition, substitution trends are evaluated and the challenges encountered were proposed from relevant research articles.

Finally, in section 5, future prospects are proposed in order to tackle criticality and recycling challenges in order to improve and scale-up LREEs recycling and to tackle the substitution challenges of LREEs in selected applications in a sustainable manner. The structure of this review article is shown in Fig. 4.
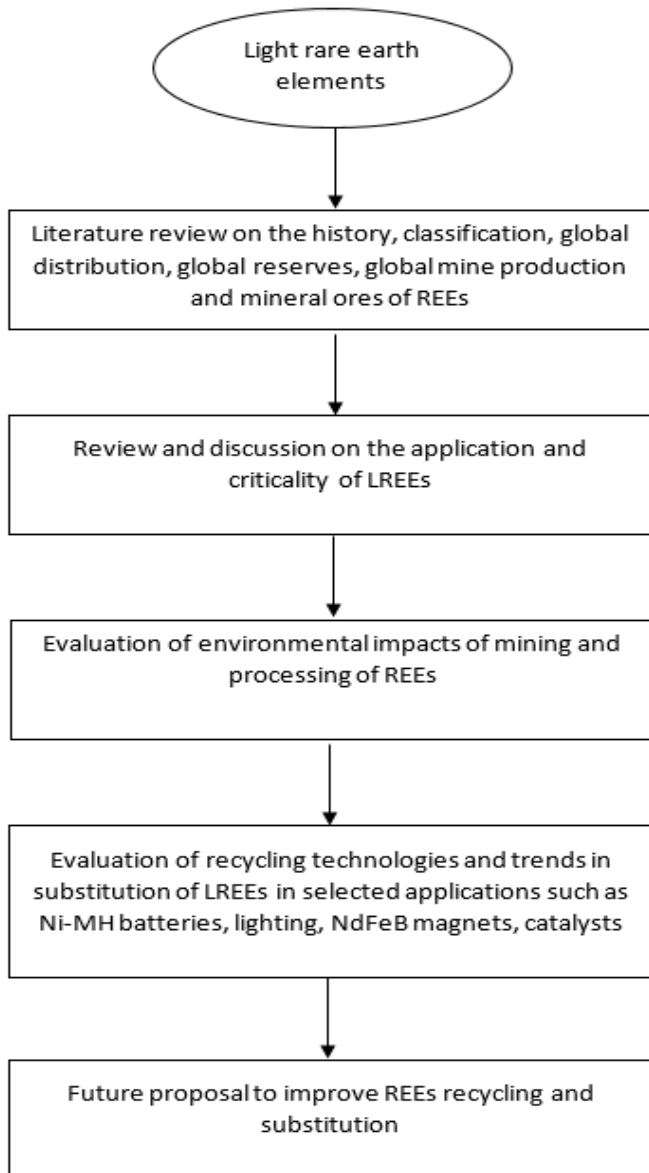
Fig. 4. Structure of the review article.

2. Applications, criticality and environmental impacts of LREEs mining and processing

2.1. Applications of LREEs

Table 1 and Table 2 highlight the application areas of LREEs and the percentage of LREEs consumption in various applications, respectively.

Table 1. LREEs in selected applications.

Application	LREEs
Magnets	Praseodymium, neodymium, samarium
Batteries	Lanthanum, cerium, neodymium, praseodymium
Lightning	Lanthanum, cerium, europium
Fluid catalytic cracking	Lanthanum, cerium
Automotive catalyst	Lanthanum, cerium, neodymium, praseodymium
Steam reforming catalyst	Lanthanum, cerium

NdFeB magnets have excellent magnetic properties, high productivity, high-energy efficiency and affordability (Svoboda, 2004). Products containing NdFeB magnets include computer hard disk drive, speakers, mobile phones, electric and hybrid vehicles, and wind turbines, air conditioning compressors, fans etc. (Constantinides, 2012) Phosphors are very important in lighting applications because of their exceptional qualities which include, emitted colour determination (Lakshmanan, 2008), high efficiency, excellent luminous intensity, low energy consumption, and durability (Shinde et al., 2012). They are also environmentally friendly, have high thermal stability, and are non-toxic (Pawade et al., 2015). Phosphors containing rare earths ions are preferred for use in lighting applications because they can emit various colours of the visible light spectrum (Wang et al., 2011). Ni-MH batteries have high energy density of up to 80 Wh/Kg, long life cycles of up to 1000 cycles, no memory effect, and free maintenance (Zhan et al., 1999). Ni-MH batteries are applicable in renewable energy technologies such as hybrid and electric vehicles (Diouf and Pode, 2015) and portable electronics such as power tools (Fernandes et al., 2013). Rare earth oxides (REOs), particularly cerium oxide and lanthanum oxide are very important in the area of catalysis and in well-established catalyst applications like fluid catalytic cracking, automotive and flue gas emission abatement systems, and in hydrogen production via steam reforming (Trovarelli, 2002; Umicore, 2018). The properties of REO catalysts that make them unique are high selectivity, activity, and stability. Cerium oxide in particular, has the following desirable characteristics: (I) improved oxygen storage capability, (II) high thermal stability, (III) ease of impregnation in certain elements, e.g. alumina, (IV) compatibility with many metals e.g. noble metals, and (V) dispersion enhancement of active metals on support. (Trovarelli, 2002; Zhan et al., 2014)

Table 2. REEs (%) consumption in various application (Roskill, 2016).

Application	REEs consumption (%)
Catalysts	24/Fluid cracking catalysts (18%) Automotive catalysts (5%)
Magnets	23
Polishing	12
Metallurgy	8
Batteries	8
Glass	7
Ceramics	6
Phosphors	2
Others	10

2.2. Criticality of LREEs

LREEs belongs to a class of metals known as 'critical metals'. Critical metals have their title because they are of enormous importance globally which makes them easily susceptible to future scarcity, they are highly demanded, their substitutes are not readily available, and they have low recycling rate (Gunn, 2013). Grandell et al. (2016) described critical metals as those metals that are important and crucial for green energy technologies. Critical metals can also be described as metals that are prone to global access and supply risks (Mishra, 2015). The supply risks can be caused by factors such as geological availability, political factors, and the evolution of technology (Gunn, 2013).

Criticality of LREEs can be identified through various factors and indicators such as the environmental risks associated with the extraction, mining, and processing of LREEs, the supply risks, the economic importance associated with the individual LREEs, and the demand (Deboer and Lammertsma, 2013; Guyonnet et al., 2015). One major concern associated with critical metal is the potential impact that can be created by supply disruptions particularly in countries that solely depend on imported metals (Gunn, 2013). The modern world has come to depend on these critical in maintaining and improving its lifestyle. Any disruption to this lifestyle could be very detrimental. In 2017, the European Commission listed the LREEs cerium, lanthanum, neodymium, and praseodymium as one of twenty-seven critical raw materials (European Commission, 2017). It is worth noting that the criticality of some elements can alter after a period, this can be caused by factors like the emergence of substitutes, sudden increase or decrease in demand, and market response (U.S. Department of Energy, 2011).

2.3. Environmental impacts of LREEs mining and processing

LREEs obtained from primary sources affect the environment during mining and processing stages causing concern. High amounts of energy and water are consumed during the extraction and processing of LREEs (Nassar et al., 2015). Waste rock stockpiles contain radioactive elements uranium and thorium. Separation of LREEs from these radioactive elements leads to health and environmental impacts and added processing costs (Golev et al., 2014). Open pit mines if exposed to rainwater will pollute the environment with radioactive and toxic substances (Schüler et al., 2011). Mine tailings also constitute to the negative environmental impacts of LREEs in both short and long terms. Mine tailings are composed of wastewater and chemicals from the flotation process. Incidents of severe pollution of underground and surface waters, contamination of soils by heavy metals, and toxic chemicals have been documented for example in China. (Golev et al., 2014) LREEs processing consumes large amounts of chemicals and water, as well as high amounts of energy, e.g. large amounts of salts are generated during the chemical processing of LREEs (De Lima, 2016). The detrimental effects of LREEs mining and processing, as well as the high cost of remediation of contaminated land, can be prevented. To do this, recycling, substitution, and effective regulatory policies would reduce significantly the need to mine LREEs.

3. Recycling

The recycling of LREEs has two forms, (1) post-consumer recycling, which entails recycling LREEs from the end-of-life consumer products and (2) pre-consumer recycling involving recycling performed on manufacturing scrap (Panayotova and Panayotov, 2012). This review focuses on post-consumer recycling of LREEs. For metals, recycling usually comprises of three steps (Fig. 5), namely a collection step, a pre-processing step and an end-processing step. In the collection step, the end-of-life products are collected and cleaned, and the pre-processing step focuses on the physical dismantling and separation of the end-of-life products. (Reck and Graedel, 2012; Zakotnik et al., 2009). The end-processing step focuses on the use of metallurgical techniques to recover LREEs (Hagelüken et al., 2016).
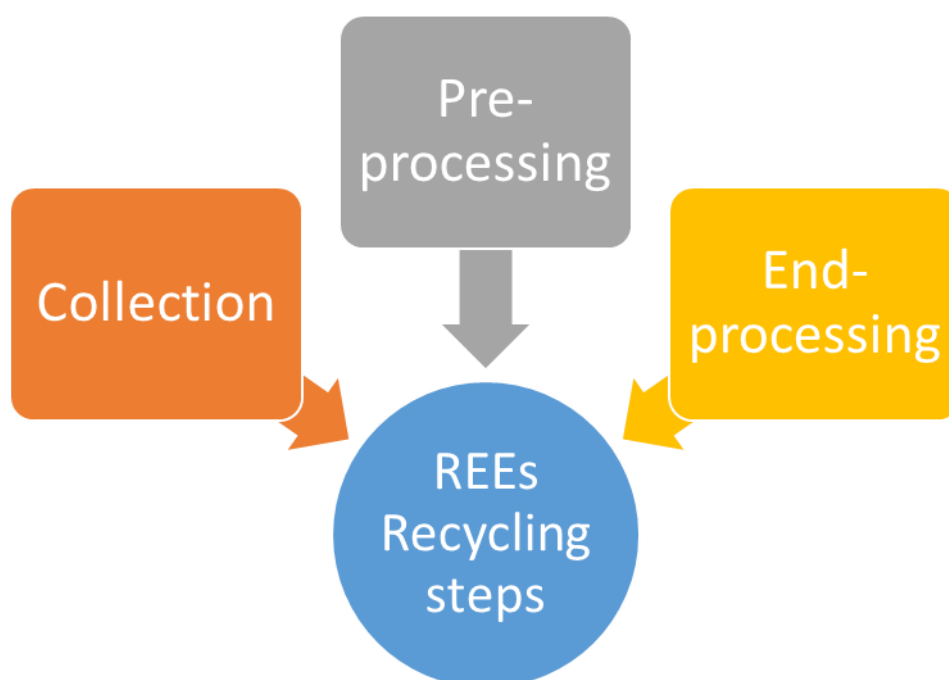

Fig. 5. LREEs recycling steps (Adapted from: Hagelüken et al., 2016; Reck and Graedel, 2012).

There are various recycling techniques used to recover LREEs from secondary sources, each of these having its advantages and disadvantages. Factors such as the LREEs composition, type of flow and chemical complexity can be used to decide what recycling technique to use for a particular flow (Tunsu et al., 2015). Hydrometallurgical and pyrometallurgical techniques are the two most commonly used LREEs recycling techniques (Gupta and Mukherjee, 1990).

In the hydrometallurgical technique, selective separation of LREEs occurs whereby the LREEs bearing alloys are dissolved in large amounts by powerful mineral acids (leaching) and precipitated to form fluorides, oxalates or double sulfates. Leaching can also be performed using microorganisms in an environmentally friendly process known as bioleaching. Hydrometallurgical techniques include (1) leaching, (2) solvent extraction, (3) ion exchange, and (4) precipitation. (Brierley and Brierley, 2013; Gupta and Mukherjee, 1990; Müller and Friedrich, 2006)

The pyrometallurgical technique uses very high temperatures to recover metals (EPA, 1994). The pyrometallurgical technique utilizes high temperatures and it is mostly suited for end-of-life products having high concentrations of LREEs. Pyrometallurgical techniques include (1) roasting, (2) liquid metal extraction, (3) direct melting, (4) gas-solid and solid-liquid, (5) glass slag process, (6) electrochemical processing, (7) molten salt extraction, (8) molten slag extraction, and (9) gas-phase extraction (Firdaus et al., 2016; Gupta and Krishnamurthy, 1992; King et al., 2016; Parkinson, 1987; Yang et al., 2016)

Recycling of LREEs is a very complex procedure and has several challenges restricting its progress. The challenges encountered are (1) inadequate recycling technology, (2) very little or no recycling incentives, (3) small amount of LREEs in products, (4) lack of efficient dismantling, separation, and collection systems, (5) declining LREEs prices, (6) lack of supply shortages, (7) long useful life period (up to 10-20 years) of some LREEs bearing product (e.g. electric vehicles and wind turbines containing large permanent magnets), (8) lack of effective governmental policies, and (9) air transportation of magnetic materials. (Dutta et al., 2016; Eggert et al., 2016; Krishnamurthy and Gupta, 2015; Panayotova and Panayotov, 2012; Tunsu et al., 2015)

Despite the challenges encountered in LREEs recycling, new advances are being made that can lead to more sustainable, effective and selective LREEs recycling technologies, particularly in the area of permanent magnets (Dupont and Binnemans, 2015).

3.1. NdFeB magnets

End-of-life NdFeB magnets contain the highest amount of recyclable LREEs (Binnemans et al., 2013). There is a huge potential for recycling neodymium from the end-of-life NdFeB magnets found in computer hard disk drives because approximately 600 million of these hard disk drives are produced yearly. In addition to this, there is an already existing efficient collection system for obtaining the end-of-life hard disk drives. (Firdaus et al., 2016) Recycling of the end-of-life hard disk drives containing NdFeB magnets starts by physically dismantling them from their computer or laptop enclosures, followed by shredding. In most cases, waste electrical and electronic equipment (WEEE) containing end-of-life NdFeB magnets are shredded directly without pre-dismantling them because of their relatively small size (Yang et al., 2014). The shredding is done to erase all data on the hard disk drives (Walton et al., 2015). After shredding, thermal demagnetization follows. Demagnetization of the already shredded hard disk drives is done by heating at 312 ℃. Size reduction is performed after this step by using a mechanical grinder and crusher to separate the NdFeB magnet bits from the hard disk drive. Finally, screening is performed to achieve improved end-of-life magnetic materials. (Abrahami et al., 2015) Hitachi developed a hard disk drive dismantling machine to replace the time-consuming manual/physical dismantling process with the use of a rotational drum. The hard disk drive is subjected to vibration and mechanical shocks, which

leads to the separation of NdFeB magnets from the hard disk drives and the demagnetization of the permanent magnet (Baba et al., 2013; Hitachi, 2010).

Researchers at the Critical Materials Institute (CIM) in conjunction with Ames laboratory have developed an award-winning novel process of recycling of LREEs from shredded hard disk drives containing NdFeB magnets. In this process, the end-of-life NdFeB magnets are dissolved in an acid-free water-based solution. The recovered LREEs had greater than 99% purity and they have been successfully reused in making new magnets (AMES laboratory, 2018).

Jakobsson et al. (2016) demonstrated a direct melting process for the recovery of rare earth oxides from NdFeB magnets contained in the end-of-life ferrous electronic materials. In this technique, the NdFeB magnets were melted at a very high temperature of 1600 $^{\circ}$C for 1 hour and the temperature was kept constant for an additional hour. The subsequent slag phase contained approximately 28% of neodymium oxide and approximately 1% of praseodymium.

Hua et al. (2014) proposed a process of selective extraction with molten chlorides for the recovery of LREEs from the end-of-life NdFeB magnets. In this process, the LREEs from the end-of-life NdFeB magnets were selectively extracted into the molten chloride salts ($MgCl_2$-KCl) at a high temperature and time of 1000 $^{\circ}$C and 3 hours, respectively. Neodymium obtained was up to 86.6% through this process, while that of praseodymium was up to 89.2%. Furthermore, the extraction of neodymium and praseodymium increased with an increase in temperature and reaction time of up to 1200 $^{\circ}$C and 12 hours, respectively.

The challenges encountered when recycling NdFeB magnets from hard disk drives include: (1) difficulties encountered in identifying and removing the NdFeB magnets from the hard disk drives, (2) the re-insertion of the recycled NdFeB magnets back to its supply chain, (3) the efficient separation of the NdFeB magnets from the other materials contained in the hard disk drives, and (4) difficulties encountered during the collection and sorting of the hard disk drive. (U.S. Department Of Energy, 2015; Walton et al., 2015)

3.2. Ni-MH batteries

Recycling the end-of-life Ni-MH batteries for the recovery of cerium, lanthanum, neodymium, and praseodymium is very essential because of their huge economic potential. The number of the end-of-life vehicles containing the Ni-MH batteries was approximately 40 million globally in 2010 (Xu et al., 2016) and the total mass of LREEs present in the Ni-MH batteries was about 4.5 kg (Alonso et al., 2012). Usually, the useful life period of the Ni-MH batteries is about 2 years (Yang et al., 2014), this translates to the generation of large amounts of the end-of-life Ni-MH batteries and consequently creates the need for recycling of LREEs from this wastes. Commercially, Umicore recycles a substantial amount of the end-of-life Ni-MH batteries and the end-of-life Li-ion batteries using its UHT (ultra-high temperature) furnace located in Hoboken Belgium with an installed capacity of up to 7000 metric tons annually. Umicore with the use of the pyrometallurgical technique has successfully recovered large quantities of cerium, lanthanum, neodymium, and praseodymium with low amounts of air emissions, negligible amount of waste generated and reduced energy requirements. (Umicore, 2018) Honda in collaboration with Japan Metals & Chemicals (JMC) has begun the reuse of LREEs in spent nickel metal hydride batteries to manufacture new batteries (Forbes, 2014). Several laboratory scale studies have been successfully carried out for the recycling of cerium, lanthanum from end-of-life Ni-MH batteries.

Innocenzi and Vegliò (2012) attempted the recovery of cerium and lanthanum from end-of-life Ni-MH batteries through selective precipitation. The initial amount of LREEs cerium and lanthanum present in the ground Ni-MH batteries was both 5% w/w. Two consecutive leaching steps were

carried out to dissolve almost all of cerium and lanthanum present. 99% of LREEs lanthanum and cerium sulfates were recovered through selective precipitation with sodium hydroxide at low pH.

Meshram et al. (2016) proposed a method of leaching neodymium, cerium, and praseodymium from end-of-life Ni-MH batteries. Leaching was performed using 2 M concentration of sulphuric acid at a temperature of 348 K (approx. 75 °C) for 2 hours. This resulted in a leaching efficiency of 98.1% for neodymium, 89.4% for praseodymium, and 89.4% for cerium.

Gasser and Aly (2013) investigated a method for the recovery of neodymium (III) and lanthanum (III) from end-of-life Ni-MH batteries through an environmentally friendly synthetic adsorbent process involving leaching and solvent extraction. Leaching occurred using 2 M sulphuric acid at 25 °C for 2 hours. After leaching, separation of LREEs residue from the leaching solution occurred by filtration. This was followed by precipitation of LREEs sulfate in the filtrate by sodium hydroxide to extract a white dense precipitate of LREEs hydroxides. Subsequent neodymium (III) and lanthanum (III) were recovered by leaching with 2 M hydrochloric acid. The recovery of neodymium (III) was above 97% while that of lanthanum (III) was below 0.8%.

3.3. Lighting

Some full-scale processes perform recycling of end-of-life phosphors in lighting applications. Solvay, an international chemical company began commercial operation for recycling end-of-life phosphors from light bulbs for the recovery of cerium and lanthanum in 2012 in its France facility (Solvay, 2012).

Yang et al. (2013) investigated a method for recovering cerium and lanthanum from end-of-life fluorescent lamps by a two-step leaching process and subsequent solvent extraction using ionic liquids (ILs). In the first leaching step, 5 M concentration of sulphuric acid at 100 °C was used to leach the phosphors powders for 6 hours. After the first leaching step, lanthanum and cerium were not completely dissolved in the acidic solution, so a second leaching step was carried out. The process condition that gave the highest leaching efficiency was at 5 M concentration of sulphuric acid at 100 °C for 6 hours. This gave a leaching efficiency of approximately 90%. It was further stated that an increase in the acid concentration would further increase the leaching efficiency. After the second leaching step, an ionic liquid was used to extract lanthanum and cerium from the leachate.

Abrahami et al. (2014) proposed direct leaching, for the recovery of neodymium from NdFeB magnets contained in the end-of-life computer hard disk drives. Direct leaching of the end-of-life permanent magnet occurred with 2 M sulphuric acid at room temperature for 8 hours. The recovery of neodymium after the leaching process was more than 95 wt-%. Subsequent precipitation followed and 98.4% neodymium was obtained as double salts. It was further observed that the direct leaching approach was more efficient than the molten slag extraction and subsequent incomplete leaching approach.

Van Loy et al. (2017) explored a mechanical activation and two-step leaching processes for the efficient recovery of lanthanum and cerium from spent green phosphors using. Mechanical activation occurred using 1 mm planetary ball mill with a rotational speed of 600 rpm. Mechanical activation, when used prior to leaching, boosts the leaching process. Subsequent leaching took place using 4 M hydrochloric acid for 2 hours at room temperature. The yield of lanthanum obtained was 99% while that of cerium was 86.3%.

Reed et al. (2016) demonstrated a technique of bioleaching waste phosphor using the microorganism *Gluconobacter oxydans*. The result showed that the total leaching efficiency of LREEs lanthanum and cerium was very low at around 2%. It was suggested that the low leaching

efficiency might be attributed to the high amount of calcium phosphate present in the waste phosphors.

Tunsu et al. (2014) developed a process of recovering cerium from waste phosphors using a two-step leaching procedure. During the first step, leaching occurred using 0.5M nitric acid at around 20 ºC for 24 hours. After the first leaching step, the undissolved cerium was subsequently leached (second-step) with 4M nitric acid solution, followed by ultrasound-assisted digestion to recover cerium. Dissolution of the contaminant mercury was possible with 4M hydrochloric acid at around 20 ºC for 168 hours. It was further proposed that in order to increase the recovery efficiency for cerium and lanthanum, longer leaching time is necessary and nitric acid should be replaced by hydrochloric acid. Lanthanum, on the other hand, could not be recovered.

Wu et al. (2014) developed an environmentally friendly method that combined alkali fusion (enables the breakdown of the waste phosphor's dense form) and acid leaching processes for the recovery of cerium from the end-of-life phosphors in waste fluorescent. Leaching occurred using a mixture of hydrochloric acid and hydrogen peroxide. The process condition of the leaching was 6M of acid at 70 ºC for 2 hours. It was further discovered that both lanthanum and cerium were slightly dissolved after leaching. Calcination was carried out to increase the leaching efficiency. Calcination occurred using sodium peroxide with a temperature of 650 ºC for 15 minutes. Cerium recovered using this method was approximately 98% while lanthanum recovered was approximately 99%. The total percentage of LREEs recovered was higher that 99.5%.

Liu et al. (2014) described a method involving a combination of leaching and alkali fusion for the recovery of cerium in end-of-life green phosphors. Leaching first occurred at 60 ºC with 4M hydrochloric acid. Cerium remained undissolved after the leaching. Subsequent alkali fusion was performed to recover the cerium residue. The optimum alkali temperature by which cerium was recovered was at 800 ºC. The leaching efficiency obtained by this method was up to 71.45% of cerium.

3.4. Catalysts

In the area of catalysis, the most prominent application in which cerium oxide and lanthanum oxide are recovered is the recycling of LREEs from the fluid catalytic cracking catalysts (Innocenzi et al., 2016). In automotive exhaust gas converters, the focus is mainly on the recovery of platinum group metals mainly because of their high value and high cost (Saitoh et al., 2017). LREEs present in end-of-life automotive exhaust gas converters are usually discarded along with the processed slag (Steinlechner and Antrekowitsch, 2015).

Zhao et al. (2017) investigated a method for the recovery of lanthanum and cerium from the end-of-life fluid catalytic cracking catalysts using leaching and solvent extraction processes. The first process step, roasting occurred at 750 ºC. This was followed by leaching with 2M hydrochloric acid at 60 ºC for 2 hours. The overall leaching efficiency for lanthanum was 72.8% while that of cerium was 86.4%.

Innocenzi et al. (2016) studied a process to recover cerium and lanthanum from the end-of-life fluid catalytic cracking catalysts by the solvent extraction process. Leaching was first carried out using 2M sulphuric acid at 80 ºC for 3 hours. Leaching efficiency of cerium obtained was 82% and that of lanthanum was 90%. After leaching solvent extraction using D2EHPA in n-heptane and subsequent precipitation with oxalic acid occurred.

Nguyen et al. (2018) proposed a three-step procedure involving leaching and a two-step solvent extraction process for the recovery of lanthanum and cerium from the end-of-life fluid catalytic cracking catalysts. Leaching first occurred using 2M nitric acid at 80 ºC. Subsequent two-step solvent extraction occurred using D2EHPA and tributyl phosphate in n-octane. The extraction yield

of cerium obtained was 89% and the lanthanum extraction yield was 72%. In Table 3, the pyrometallurgical and hydrometallurgical technique related studies from the end-of-life products for cerium, lanthanum, neodymium, and praseodymium considered in this work are being summarized.

Table 3. Recovery of neodymium, cerium, lanthanum, neodymium, and praseodymium from end-of-life products using metallurgical technology.

LREEs	Technology	Application type	Process conditions	Yield	Reference
Cerium, Lanthanum	Two-step leaching Solvent extraction	End-of-life phosphors	5M, 100 ºC, 6 hours (leaching)	90% (leaching efficiency of (lanthanum, cerium)	Yang et al. (2013)
Cerium, Lanthanum	Leaching	End-of-life green phosphors	4M, 25 ºC, 2 hours (leaching)	99% (lanthanum) 86.3% (cerium)	Van Loy et al. (2017)
Cerium, Lanthanum	Bioleaching	End-of-life phosphors	-	2 % leaching efficiency (lanthanum, cerium)	Reed et al. (2016)
Cerium	Two-step leaching	End-of-life phosphors	0.5M, 20 ºC, 24 hours (leaching 1) 4M, 20 ºC, 168 hours (leaching 2)	-	Tunsu et al. (2014)
Cerium	Leaching	End-of-life phosphors	4M, 60 ºC, 4 hour	71.45% (leaching efficiency)	Liu et al. (2014)
Cerium, Lanthanum	Leaching	End-of-life phosphors	6M, 70 ºC, 2 hour (leaching) 650 ºC, 15 minutes)	98% (cerium) 99% (lanthanum)	Wu et al. (2014)
Neodymium, Praseodymium, Cerium	Leaching	End-of-life Ni-MH batteries	2M, 75 ºC for 2 hours.	98.1% (neodymium leaching efficiency), 89.4% (praseodymium leaching efficiency),	Meshram et al. (2016)

				89.4% (cerium (leaching efficiency)	
Lanthanum, Cerium	Two-step leaching Selective precipitation	End-of-life Ni-MH batteries	3M, 80 °C, 3 hours (leaching 1) 1M, room temperature, 1 hour (leaching 2)	99% (cerium, lanthanum)	Innocenzi and Vegliò (2012)
Neodymium, Lanthanum	Leaching Precipitation	End-of-life Ni-MH batteries	2M, 25 °C, 2 hours (leaching)	97% (neodymium) 0.8% (lanthanum)	Gasser and Aly (2013)
Neodymium	Leaching	End-of-life NdFeB cs	2M, room temperature, 8 hours (leaching)	95 wt-% (neodymium leaching efficiency)	Abrahami et al. (2014)
Neodymium, Praseodymium	Direct melting	Waste NdFeB magnets	1600 °C, 1 hour	28% (neodymium) 1% (praseodymium)	Jakobsson et al. (2016)
Neodymium, Praseodymium	Molten salt extraction	End-of-life NdFeB magnets	1000 °C, 3 hours	86.6% (neodymium extraction efficiency) 89.2% (praseodymium)	Hua et al. (2014)
Lanthanum, Cerium	Roasting Leaching Solvent extraction	End-of-life fluid cracking catalysts	750 °C (Roasting) 2M, 60 °C, 2 hours (leaching)	72.8% (lanthanum leaching efficiency) 86.4% (cerium leaching efficiency)	Zhao et al. (2017)
Lanthanum, Cerium	Leaching Solvent extraction	End-of-life fluid cracking catalysts	2M, 80 °C, 3 hours (leaching)	90% (lanthanum leaching efficiency) 82% (cerium leaching efficiency)	Innocenzi et al. (2016)

| Lanthanum, Cerium | Leaching Two-step solvent extraction | End-of-life fluid cracking catalysts | 2M, 80 °C, 1 hour (leaching) | 89% (cerium) 72% (lanthanum) | Nguyen et al. (2018) |

4. Substitution

Owing to the critical nature of LREEs, there is a need to provide alternatives or in other words, substitutes to replace LREEs. Substitutes of LREEs in most cases either are inferior alternatives or still undiscovered. In many applications, LREEs substitution translates to a loss in performance (European Commission, 2017). The use of substitutes can sometimes result in the creation of an entirely new product different from what would have been obtainable if LREEs were used to create the product (Gupta and Krishnamurthy, 2005). For substitution to be completely successful, it has to replace a critical element with a more abundant one while still maintaining its intended purpose (Brumme and Brumme, 2014). Nowadays, the focus in many studies is in the reduction of the quantity of LREEs used in selected applications (European Commission, 2017).

Substitution of LREEs can take place in four different ways. The first is termed element-for-element substitution also known as direct substitution. In this case, an element is substituted by another element while maintaining its fundamental properties (Smith and Eggert, 2016; U.S. Department Of Energy, 2015). Direct substitution means that the new material developed should have the same function as the substituted LREEs material (U.S. Department Of Energy, 2015). In most cases, there are no direct substitutes for LREEs materials (De Lima, 2016). For instance, in applications like phosphors requiring optical properties are often the most difficult ones to find substitutes (Gunn, 2013). In the area of catalysis, substitutes found to replace cerium in the automotive catalyst are other critical LREEs. In fluid catalytic cracking, it is difficult to find a suitable substitute to replace lanthanum (European Commission, 2017).

A second way in which substitution can occur is termed technology-for-element. Here the LREEs is used in reduced amounts using alternative manufacturing technology. Example of this type of substitution occurs in permanent magnets. In this case, the LREEs content present in it is used in reduced amount while still retaining its underlining properties The third type is grade-for-grade substitution, this happens when a manufacturer uses an alternate category of materials in place of another, in other words, one grade of material is replaced by another grade. For example, the permanent magnet grade type contained in the direct drive of a wind turbine is substituted with another grade type. (Smith and Eggert, 2016) The fourth type is system-for-system substitution (system-level substitution). Here substitution occurs when one system replaces another system (Eggert et al., 2016; Smith and Eggert, 2016). System-level substitution takes place by redesigning the system whilst performing the same function as that of the LREEs system with reduced or equivalent output (U.S. Department Of Energy, 2015). An example of system-level substitution is when direct drive containing LREEs found in wind turbines is substituted with a gearbox and an induction generator that is LREEs free (Smith and Eggert, 2016; U.S. Department Of Energy, 2015).

4.1. NdFeB magnets

Current research in NdFeB magnets substitution focuses on reducing the quantity of neodymium and praseodymium required to produce the magnets (Widmer et al., 2015). There are presently substitutes available for NdFeB magnets found in applications like electric motors, electric vehicles,

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hybrid vehicles, and wind turbines both in industrial scale and in research and development levels. Magnet-for-magnet substitution occurs when NdFeB magnets are substituted by either samarium-cobalt, ferrite or aluminum-nickel-cobalt magnets (Eggert et al., 2016; Widmer et al., 2015). Samarium-cobalt, ferrite and aluminum-nickel-cobalt magnet are potential substitutes to the NdFeB magnet. These substitutes, however, have lower energy, lower efficiency, and lower magnetic properties than the NdFeB magnet (Brumme and Brumme, 2014; Widmer et al., 2015). Larger magnets would have to be produced or the system would have to be altered to suit the substitute magnets. Samarium-cobalt is brittle and is not able to integrate well in motors; cobalt is expensive (Smith and Eggert, 2016; U.S. Department Of Energy, 2015). Samarium-cobalt magnets, however, can function at temperatures higher than NdFeB magnets and are well suited in small, high-temperature systems (Smith Stegen, 2015).

It is possible to substitute NdFeB magnets found in synchronous electric motors with electromagnets. In this case, the motor is designed differently from the one that uses NdFeB magnets (Schüler et al., 2011). Riba et al. (2016) analyzed in details substitutes to NdFeB magnets in full and hybrid electric vehicles. One plausible substitute is the squirrel cage induction asynchronous motors, which have the properties of easy to design, low maintenance and cost, good reliability and ruggedness. They, however, have the disadvantages of low operational speed control difficulties, low efficiency etc. Despite their inherent drawbacks, they have been successfully used commercially to manufacture electric cars like Mercedes Benz, Toyota, Smith Electric vehicles, Tesla S-2014 and Tazzari Zero etc. (Akatsu and Matsui, 2013; Dorrell et al., 2010).

Switched reluctance machines is another type of motor used in electric vehicles that have been designed to be free of NdFeB magnets (Riba et al., 2016). They are easy to construct, produce and design, are low-cost motors, demonstrate low inertia and possess good thermal properties (Dorrell et al., 2010; Kumar and Jain, 2014; Riba et al., 2016). Their drawbacks are; they are difficult to control, have lower efficiency, strong vibrations and acoustic noise levels (Dorrell et al., 2010).

Synchronous reluctance motors are yet another motor designed for use in electric vehicles without the need of NdFeB magnets. These motors are characterized by magnetic steel laminations (Riba et al., 2016). One advantage of synchronous reluctance motor is that it is easy to produce (Cai et al., 2014). The drawback includes lower efficiency, lower power factor and lower torque density (Carraro et al., 2013; Morimoto et al., 2014), larger torque ripple than the motors that utilize NdFeB magnets. Despite their drawbacks, they have been used commercially for industrial applications (Riba et al., 2016).

Widmer et al. (2015) explained the possibility of substitution of NdFeB magnets in traction motors of electric vehicles. It involved the use of NdFeB magnets in reduced quantities and simultaneously increasing or at least maintaining their efficiency. The most common form of this is the reduced amount of NdFeB magnets with rotor designs. Industries like BMW have commercialized the use of this method in manufacturing electric vehicles.

In direct-drive wind turbine generators, NdFeB magnets can be substituted with copper field windings (Abrahamsen et al., 2012; Goudarzi and Zhu, 2013) in a system-for-system substitution type. The benefits of NdFeB magnet direct drives are higher reliability, lower weight, and higher efficiency (Goudarzi and Zhu, 2013; Hossain and Ali, 2015) in comparison to the copper field winding direct drive. The disadvantages of the NdFeB magnet direct drives are they are more expensive, and the magnet is prone to demagnetization. Copper field windings direct drives has the advantage of being cheaper in comparison to the permanent magnet direct drives. (Goudarzi and Zhu, 2013) Its main drawback is difficulty in installation and transportation due to its large size (Goudarzi and Zhu, 2013). Direct drive turbines free of NdFeB magnets are presently applicable on

a commercial scale. Enercon uses this technology for their onshore wind turbine installation (Enercon, 2017).

4.2. Ni-MH batteries

Rechargeable batteries used in renewable energy technology are expected to have the following properties: (1) high energy efficiency, (2) fast charging time, (3) very little or no maintenance required, (4) affordable cost, (5) environmentally friendly, (6) long useful life period, and (7) low memory effect. (Armand and Tarascon, 2008; Suppo, 2006; Zeng et al., 2014). Ni-MH batteries have been successfully substituted with lithium-ion batteries (Li-ion) (Schüler et al., 2011). Li-ion batteries are nowadays the preferred battery choice for use in portable devices like mobile phones and personal computers. They are also the dominant battery for use in electric and hybrid vehicles and in plug-in vehicles. A Li-ion battery has higher gravimetric energy, longer useful life period, no maintenance compared to Ni-MH batteries (Battery University, 2019; Sonoc et al., 2015). On the other hand, the end-of-life nickel-cadmium batteries are not a plausible substitute for Ni-MH batteries mainly because nickel and cadmium contains carcinogenic agents which are harmful to human health (Hazotte et al., 2015).

4.3. Lighting

The disadvantage of cerium and lanthanum containing phosphors used in lighting applications is the high production cost as well as the criticality (European Commission, 2017; Ogi et al., 2008). Phosphor substitutes that are completely free of cerium and lanthanum are; (I) semiconductor quantum dots, (II) phosphors comprising of oxynitride and nitride compounds, (III) carbon-based boron oxynitride compounds, (IV) transition metal ions and (V) halophosphates. Semiconductor quantum dots are considered unsuitable substitutes because of their very toxic nature, low luminosity and reproducibility difficulties. (Mao et al., 2015; Yang et al., 2012) Phosphors comprising of oxynitride and nitride compounds emit only white light. Carbon-based boron oxynitride compounds are cheaper than cerium and lanthanum (Ogi et al., 2008). On the negative side, carbon-based boron oxynitride compounds have very low quantum efficiency and weak emission peak in comparison to LREEs phosphors (Yang et al., 2012). For transition metal ions, potential substitutes are manganese ions, copper ions etc. Copper ions are the closest and most promising substitutes to cerium and lanthanum in phosphors (Mao et al., 2015).

Halophosphate phosphors are phosphors that do not contain cerium and lanthanum, they emit only white light and have no colour variant unless they are mixed with LREEs (McKeag and Ranby, 1949). Polikarpov et al. (2015) demonstrated the use of manganese based aluminum nitride (AlN:Mn2^+) in creating a plausible substitute to rare earth phosphors. The obtained quantum yield displayed very high photoluminescence. Boonsin et al. (2015) described a novel method for substituting cerium and lanthanum in phosphors using luminescent compounds based on 2,6-dimethyl-4-pyrone and Schiff based ligand. The phosphors examined consisted of one metallo-organic and two purely inorganic ligands. The result demonstrated that the rare earth free phosphors covered wide absorption areas in ultraviolet (UV) and blue regions

4.4. Catalysts

In fluid catalytic cracking, research has been carried out to substitute cerium with critical HREEs yttrium. Liu et al. (2016) attempted the substitution of cerium ion in cerium-Y zeolite with yttrium-Y zeolite. The result showed that the yttrium-Y zeolite catalyst exhibited higher conversion and light oil yield than the cerium-Y zeolite. In automotive exhaust gas catalysts, potential substitutes to cerium and lanthanum as supports are gamma alumina and aluminum phosphate (Beppu et al.,

2018; Zheng et al., 2014). However, none of these substitutes have the same excellent oxygen storage capability that cerium possesses. Beppu et al. (2018) reported an iron-rich $Pd/Sr_3Fe_{1-x}O_{3-\delta}$ automotive exhaust catalyst with a perovskite derived structure that exhibited comparable oxygen storage capability with the cerium catalyst.

For steam reforming reactions, cerium oxide and lanthanum oxide supports have been substituted with aluminum oxide and zirconia. These substitutes have comparable or sometimes higher catalytic activity than the cerium and lanthanum-based catalysts. (Kourtelesis et al., 2015; Liguras et al., 2003) Cerium oxide because of its unique redox properties and its powerful metal-support interaction is still very much preferred in steam reforming catalysts (Hou et al., 2015). Table 4 gives a summary of various substitutes for cerium, lanthanum, and neodymium, their advantages, and disadvantages for their application type considered in this work.

Table 4. Summary of substitutes for cerium, lanthanum and neodymium and their advantages and disadvantages in selected applications

Application type	LREEs product	Substitute product	Substitute Advantages	Substitute Disadvantages
Batteries	Ni-MH batteries	Li-ion batteries Nickel-cadmium (NiCd) batteries	• Li-ion batteries have higher energy efficiency than Ni-MH batteries • Li-ion batteries have fast charge time equal to Ni-MH batteries (2-4 hrs) • Li-ion batteries have no maintenance requirement, on the other hand, Ni-MH batteries require maintenance every 2-3 months • Li-ion batteries have a much slower discharging cycle than Ni-MH batteries • Li-ion batteries are non-hazardous	• Li-ion batteries are more expensive than Ni-MH batteries • NiCd batteries are toxic • NiCd batteries have a lower power density than Ni-MH batteries • NiCd batteries have a slower charge time Ni-MH batteries

			• Li-ion batteries have longer service life than Ni-MH batteries • Li-ion batteries have lower memory effect than Ni-MH batteries	
Phosphors	Phosphors containing cerium and lanthanum	• Semi-conductor quantum dots phosphors • Phosphors containing carbon oxynitride and nitride compounds • Carbon-based boron oxynitride • Phosphors containing transition metal ions • Phosphors containing luminescent compounds (metallo-organic, pure inorganic ligand) • Halophosphate phosphors	• Phosphors containing transition metal ions have a lower cost than phosphors containing cerium and lanthanum • Phosphors containing boron carbon oxynitride are cheaper than cerium and lanthanum-based phosphors	• Semi-conductor quantum dots phosphors have high toxicity, low luminosity and it is difficult to reproduce • Lower quality (halophosphate) than phosphors containing cerium and lanthanum • No colour variant emits only white light (halophosphate) • Phosphors containing luminescent compounds has thermal instability
Magnet	NdFeB magnet	• Ferrite magnet • Samarium-cobalt magnet • Electromagnet • Aluminium-nickel cobalt	Samarium-cobalt magnet can function at higher temperatures than NdFeB magnet	• Lower coercivity • Lower magnetic energy • Lower efficiency • Lower magnetic properties than that of the NdFeB magnet

				• Samarium-cobalt magnet is brittle
				• Samarium-cobalt magnet is unable to integrate well in motors
				• Cobalt is expensive
Electric and Hybrid Vehicles	NdFeB magnet	• Squirrel cage induction asynchronous motors	• Easy design • Lower maintenance required • Lower cost • Reliability and rugged	• Lower operational speed • Lower efficiency • Speed control problems than that of the NdFeB magnet
		• Switched reluctance machines	• Easy to construct, design and produce • Low cost • Low inertia • Good thermal properties	• Control problems • Lower efficiency • Strong vibrations • Acoustic noise levels
		• Synchronous reluctance motors	• Easy to produce	• Lower efficiency • Lower power factor • Lower torque density • Large torque ripple than that of the NdFeB magnet
Wind turbine generators	NdFeB magnets	• Copper field windings (electrically excited) drives	• Low cost	• Installation difficulties • Its large size creates problems in transporting it
Fluid catalytic cracking	Cerium-Y zeolite	Yttrium-Y zeolite	• Higher conversion than cerium-based Y zeolite • Higher thermal stability than cerium-based Y zeolite	• Yttrium-Y zeolite has lower acid sites than the cerium-based Y zeolite
Automotive exhaust catalysts	Cerium/lanthanum-based catalysts	• Iron-rich $Pd/Sr_3Fe_{1-x}O_3{-}_\delta$	• Comparable oxygen storage capabilities as cerium and lanthanum-based	• Higher cost than the cerium/lanthanum-based catalysts

Steam reforming	Cerium oxide	Zinc oxide	catalysts • Higher catalytic activity	• Zinc oxide has lower oxygen storage capabilities than the cerium oxide catalyst

5.   Future strategies for sustainability of LREEs recycling and substitution

The United Nations Brundtland Commission defined sustainable development as "development that meets the needs of the present without compromising the ability of future generations to meet their own needs" (United Nation, 1987). Sustainability cuts across environmental, social and economic dimensions, which must be balanced in order to achieve the goals (Azapagic and Perdan, 2000).

5.1.   Sustainability benefits of LREEs recycling

From the environmental point of view, recycling of LREEs from end-of-life products is more sustainable than the extraction of virgin ore from the earth crust. The extraction of earth's natural resources is a non-renewable process and leads to depletion of finite resources. Additionally, environmental pollutants such as radioactive elements and dust are generated during the mining and processing stages. Noise pollution, water and soil contamination, increased carbon footprint and energy consumption are also the negative effect of producing LREEs from primary sources. Moreover, end-of-life LREEs bearing products dissipated into landfills makes them inaccessible to future generation. Recycling on the other hand, does not lead to the depletion of earth's natural resources, thereby conforming to the Sustainability goal of preserving earth's resources for future generation. Recycling also has lesser environmental impacts. It eliminates the negative environmental impacts of LREEs mining and processing. Finally, it prevents the disposal of end-of-life LREEs bearing products into landfills by utilizing them.

From a social point of view, recycling of LREEs from end-of-life products is a more sustainable option than the extraction of virgin ore from the earth crust. REEs mining and processing is harmful to both humans and aquatic health. The radioactive elements produced during mining operation are carcinogenic. In addition, the large amount of salts and chemicals generated during the processing of LREEs could lead to the contamination of both surface and groundwaters, which in turn could negatively affect humans and aquatic life. On the other hand, increased recycling of LREEs could lead to an increased number of recycling centers, which could further lead to increased job creation, which is a social incentive that is beneficial to human well-being.

From an economic point of view, with the declining prices of LREEs and lack of supply shortages, LREEs from primary sources is currently the more preferred option. On the other hand, due to the several challenges of LREEs recycling such as the inadequacy in recycling technologies, lack of enforceable governmental policies and low LREEs concentration in the end-of-life LREEs bearing products, the recycling of LREEs currently has lesser economic benefits. For recycling to be economically beneficial, a balance must be formed between the cost of recycling and the resulting benefits. The cost of recycling should not be too expensive and should be profitable for all stakeholders involved. This could lead to increased commercial recycling of LREEs. One of the major challenges of LREEs recycling is the lack of an efficient collection system. One way to address this challenge is to educate the public as well as concerned stakeholders on the benefits of

recycling and its implication on future generations. Recycling programs are one way in which education of LREEs recycling can be provided. There should be an increased focus on research and development to improve the already existing collection system. Having several collection centers and making them easily accessible is also beneficial. Finally, there should be effective governmental regulations and incentives on the LREEs collection systems. The government can, e.g. introduce take-back programs with financial incentives for the end-of-life LREEs products as well as enforceable penalties to defaulters of governmental policies. If done correctly, recycling can also lead to the reduction of dependence by non-LREEs producing countries on LREEs producing countries which is beneficial for long-term sustainable development.

5.2. Substitution strategies

Substitution if done properly is a plausible way to reduce significantly the demand for LREEs. Research and development into potential LREEs substitutes as well as the development of new technology for substitution would go a long way in addressing the criticality concerns of LREEs. Direct substitution of LREEs with other materials in different applications has recorded very little success. Several attempts on direct substitution have either led to a lowered performance of the product or resulted in the substitution of critical LREEs by other critical LREEs. Technology-for-element substitution is more plausible than the direct substitution because it involves the use of LREEs in reduced amounts using alternate technologies. It is, however, not ideal because it still makes use of the critical LREEs. In grade-for-grade substitution type, the challenge faced is finding a similar or higher-grade material than LREEs.

The system-level substitution is the broadest and the most versatile form of substitution that offers flexibility. It has been applied successfully in wind turbines and in electric vehicles. More research and development is needed to extend this substitution type to other LREEs product areas.

Because LREEs are currently cheap and established in many application areas, the development of suitable substitutes has been very slow. To combat this challenge, government intervention is needed in two ways; (I) provision of funds for research and development to finding suitable LREEs substitute, and to promote existing substitutes for improvement in performance, and (II) reduction of re-engineering costs encountered in substitution.

6. Conclusion

Criticality and environmental challenges of obtaining LREEs from primary sources have led to the emergence of several recycling techniques and substitution methods. Production of LREEs from virgin ore causes environmental degradation. Mining activities involving LREEs have generated environmental concerns globally. Mine tailings have a negative impact on the environment. In addition, waste rock stockpiles contain radioactive elements that are carcinogenic to humans. Processing of LREEs consumes large amounts of energy and produces wastewaters. Recycling and substitution are essential for ensuring the sustainability of LREEs resources thereby ensuring that they are conserved for the future generations. Recycling of post-consumer end-of-life LREEs bearing products can take place in three steps, namely the collection step, pre-processing step and end-processing step. Commercial recycling has been slowly developing mainly because of the low concentration of LREEs present in some end-of-life products. That makes recycling more expensive than the extraction from virgin ores. The long useful life period (up to 10-20 years) of some LREEs bearing product e.g. electric vehicles and wind turbines containing large NdFeB magnet is another reason for the slow development of commercial LREEs recycling. The lack of efficient collection

system and technological barriers has also been cited for the slow development of LREEs on the commercial scale. However, recycling of LREEs on the laboratory scale has been advancing steadily. Recycling techniques such as hydrometallurgical and pyrometallurgical processing have been used to recover LREEs successfully from the end-of-life products with a high purity of up to 99% in some cases. The end-of-life NdFeB magnets contain the highest amount of recyclable LREEs when compared to other end-of-life LREEs products. If recycling of LREEs is adequately harnessed, it can complement the primary sources of LREEs. For recycling to improve, action needs to be taken by all stakeholders including the involved government.

Substitutes to LREEs in many cases are either inferior or still undiscovered. In many applications, LREEs substitution translates to a loss in performance. For substitution to be completely successful, it has to replace the critical LREEs with a more abundant element while still maintaining its intended purpose. Nowadays, the focus of many researches is in the reduction of the quantity of LREEs used in certain applications. Substitution of LREEs can be carried out in four ways namely, direct substitution, technology-for-element substitution, grade-for-grade substitution, and system-level substitution. Plausible system-level substitutes have been found in applications like electric vehicles and wind turbines. A major challenge of substitution is that LREEs are cheap, very efficient and established in many application areas. To combat this challenge, government interventions are needed in two ways; (I) provision of funds for research and development to finding suitable LREEs substitute, and to promote existing substitutes for improvement in performance, and (II) reduction of re-engineering costs encountered in substitution.

In conclusion, intensive research should further be carried out to build efficient and effective recycling and substitution schemes.

Acknowledgments

References

Abrahami, S.T., Xiao, Y., Yang, Y., 2015. Rare-earth elements recovery from post-consumer hard-disc drives. Miner. Process. Extr. Metall. 124, 106–115.

Abrahamsen, A.B., Magnusson, N., Jensen, B.B., Runde, M., 2012. Large superconducting wind Turbine generators. Energy Procedia 24, 60–67.

Akatsu, K., Matsui, N., 2013. New trend of motor technology for automobiles - Introduction and overview, in: 2013 IEEE ECCE Asia Downunder.

Alonso, E., Wallington, T., Sherman, A., Everson, M., Field, F., Roth, R., Kirchain, R., 2012. An Assessment of the Rare Earth Element Content of Conventional and Electric Vehicles. SAE Int. J. Mater. Manuf. 5, 473–477.

AMES laboratory, 2018. Rare-earth magnet recycling tech wins innovation award [WWW Document]. URL https://www.ameslab.gov/news/news-releases/rare-earth-magnet-recycling-tech-wins-innovation-award (accessed 4.30.19).

Armand, M., Tarascon, J.-M., 2008. Building better batteries. Nature 451, 652–657.

Azapagic, A., Perdan, S., 2000. Indicators of sustainable development for industry: A general framework. Process Saf. Environ. Prot. 78, 243–261.

Baba, K., Hiroshige, Y., Nemoto, T., 2013. Rare-earth magnet recycling. Hitachi Rev. 62, 452–455.

Battery University, 2019. Advantages and limitations of the Different Types of Batteries - Battery University [WWW Document]. URL

https://batteryuniversity.com/learn/archive/whats_the_best_battery (accessed 4.30.19).

Beppu, K., Demizu, A., Hosokawa, S., Asakura, H., Teramura, K., Tanaka, T., 2018. Pd/SrFe 1– x Ti x O 3−δ as Environmental Catalyst: Purification of Automotive Exhaust Gases. ACS Appl. Mater. Interfaces 10, 22182–22189.

Binnemans, K., Jones, P.T., Blanpain, B., Van Gerven, T., Yang, Y., Walton, A., Buchert, M., 2013. Recycling of rare earths: A critical review. J. Clean. Prod. 51, 1–22.

Boonsin, R., Chadeyron, G., Roblin, J.P., Boyer, D., Mahiou, R., 2015. Development of rare-earth-free phosphors for eco-energy lighting based LEDs. J. Mater. Chem. C 3, 9580–9587.

Brierley, C.L., Brierley, J.A., 2013. Progress in bioleaching: part B: applications of microbial processes by the minerals industries. Appl. Microbiol. Biotechnol. 97, 7543–7552.

British Geological Survey, 2011. Rare earth elements [WWW Document]. URL https://www.bgs.ac.uk/downloads/start.cfm?id=1638 (accessed 4.26.19).

Brumme, A., Brumme, A., 2014. Market analysis of rare earth elements, in: Wind Energy Deployment and the Relevance of Rare Earths. Springer Gabler, p. 108.

Cai, H., Guan, B., Xu, L., 2014. Low-cost ferrite PM-assisted synchronous reluctance machine for electric vehicles. IEEE Trans. Ind. Electron. 61, 5741–5748.

Carraro, E., Degano, M., Bianchi, N., 2013. Permanent magnet volume minimization in permanent magnet assisted synchronous reluctance motors, in: 2013 8th International Conference and Exhibition on Ecological Vehicles and Renewable Energies, EVER 2013. pp. 1–4.

Constantinides, S., 2012. The Demand for Rare Earth Materials in Permanent Magnets [WWW Document]. Rare Earths. URL https://www.magmatllc.com/PDF/Demand for rare earth materials in permanent magnets_Constantinides_120409.pdf (accessed 1.5.19).

De Lima, I.B., 2016. Rare Earths Industry and Eco-management: A Critical Review of Recycling and Substitutes, in: De Lima, I.B., Filho, W.L. (Eds.), Rare Earths Industry: Technological, Economic, and Environmental Implications. Elsevier, pp. 293–304.

Deboer, M.A., Lammertsma, K., 2013. Scarcity of rare earth elements. ChemSusChem 6, 2045–2055.

Diouf, B., Pode, R., 2015. Potential of lithium-ion batteries in renewable energy. Renew. Energy 76, 375–380.

Dorrell, D.G., Knight, A.M., Popescu, M., Evans, L., Staton, D.A., 2010. Comparison of different motor design drives for hybrid electric vehicles, in: 2010 IEEE Energy Conversion Congress and Exposition, ECCE 2010 - Proceedings. pp. 3352–3359.

Dupont, D., Binnemans, K., 2015. Rare-earth recycling using a functionalized ionic liquid for the selective dissolution and revalorization of Y2O3:Eu3+ from lamp phosphor waste. Green Chem. 17, 856–858.

Dutta, T., Kim, K.H., Uchimiya, M., Kwon, E.E., Jeon, B.H., Deep, A., Yun, S.T., 2016. Global demand for rare earth resources and strategies for green mining. Environ. Res. 150, 182–190.

Eggert, R.G., Wadia, C., Anderson, C., Bauer, D., Fields, F., Meinert, L., Taylor, P., 2016. Rare Earths: Market Disruption, Innovation, and Global Supply Chains. Annu. Rev. Environ. Resour. 41, 199–222.

Enercon, 2017. Production for ENERCON Impressive production network [WWW Document]. URL https://www.enercon.de/en/company/production/ (accessed 5.2.19).

EPA, 1994. Handbook: Recycling and Reuse of Material Found on Superfund Sites. Diane Publishing Co.

European Commission, 2017. Communication from the commission to the European parliament, the council, the European economic and social committee and the committee of the regions: on the 2017 list of Critical Raw Materials for the EU [WWW Document]. URL https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX:52017DC0490 (accessed 5.2.19).

Fernandes, A., Afonso, J.C., Dutra, A.J.B., 2013. Separation of nickel(II), cobalt(II) and lanthanides from spent Ni-MH batteries by hydrochloric acid leaching, solvent extraction and

precipitation. Hydrometallurgy 133, 37–43.

Firdaus, M., Rhamdhani, M.A., Durandet, Y., Rankin, W.J., McGregor, K., 2016. Review of High-Temperature Recovery of Rare Earth (Nd/Dy) from Magnet Waste. J. Sustain. Metall. 2, 276–295.

Forbes, 2014. Rare Earth Recycling Takes On New Luster [WWW Document]. URL https://www.forbes.com/sites/heatherclancy/2014/02/25/rare-earth-recycling-takes-on-new-luster/#1bb26b01543d (accessed 5.2.19).

Gambogi, J., 2016. Rare earths. U.S. Geological Survey, Mineral Commodity Summaries.

Gasser, M.S., Aly, M.I., 2013. Separation and recovery of rare earth elements from spent nickel-metal-hydride batteries using synthetic adsorbent. Int. J. Miner. Process. 121, 31–38.

Golev, A., Scott, M., Erskine, P.D., Ali, S.H., Ballantyne, G.R., 2014. Rare earths supply chains: Current status, constraints and opportunities. Resour. Policy 41, 52–59.

Goudarzi, N., Zhu, W.D., 2013. A review on the development of wind turbine generators across the world. Int. J. Dyn. Control 1, 192–202.

Grandell, L., Lehtilä, A., Kivinen, M., Koljonen, T., Kihlman, S., Lauri, L.S., 2016. Role of critical metals in the future markets of clean energy technologies. Renew. Energy 95, 53–62.

Gunn, G., 2013. Critical Metals Handbook, Critical Metals Handbook. John Wiley & Sons, Ltd.

Gupta, C.K., Krishnamurthy, N., 2005. Extractive Metallurgy of Rare Earths. CRC Press.

Gupta, C.K., Krishnamurthy, N., 1992. Extractive metallurgy of rare earths. Int. Mater. Rev. 37, 197–248.

Gupta, C.K., Mukherjee, T.K., 1990. Hydrometallurgy in extraction processes. CRC Press.

Guyonnet, D., Planchon, M., Rollat, A., Escalon, V., Tuduri, J., Charles, N., Vaxelaire, S., Dubois, D., Fargier, H., 2015. Material flow analysis applied to rare earth elements in Europe. J. Clean. Prod. 107, 215–228.

Hagelüken, C., Lee-Shin, J., Carpentier, A., Heron, C., 2016. The EU Circular Economy and Its Relevance to Metal Recycling. Recycling 1, 242–253.

Haskin, L.A., Frey, F.A., 1966. Dispersed and Not-so-rare earths. Science. 152, 299–314.

Hazotte, C., Leclerc, N., Diliberto, S., Meux, E., Lapicque, F., 2015. End-of-life nickel-cadmium accumulators: Characterization of electrode materials and industrial Black Mass. Environ. Technol. 36, 796–805.

Hitachi, 2010. Hitachi Develops Recycling Technologies for Rare Earth Metals [WWW Document]. URL http://www.hitachi.com. (accessed 5.2.18).

Hossain, M.M., Ali, M.H., 2015. Future research directions for the wind turbine generator system. Renew. Sustain. Energy Rev. 49, 481–489.

Hou, T., Zhang, S., Chen, Y., Wang, D., Cai, W., 2015. Hydrogen production from ethanol reforming: Catalysts and reaction mechanism. Renew. Sustain. Energy Rev. 44, 132–148.

Hua, Z., Wang, J., Wang, L., Zhao, Z., Li, X., Xiao, Y., Yang, Y., 2014. Selective extraction of rare earth elements from NdFeB scrap by molten chlorides. ACS Sustain. Chem. Eng. 2, 2536–2543.

Innocenzi, V., De Michelis, I., Ferella, F., Veglio, F., 2016. Rare earths from secondary sources: profitability study. Adv. Environ. Res. 5, 125–140.

Innocenzi, V., Vegliò, F., 2012. Recovery of rare earths and base metals from spent nickel-metal hydride batteries by sequential sulphuric acid leaching and selective precipitations. J. Power Sources 211, 184–191.

International Union of Pure and Applied Chemistry, 2016. IUPAC Periodic Table of the Elements [WWW Document]. URL https://iupac.org/wp-content/uploads/2018/12/IUPAC_Periodic_Table-01Dec18.pdf (accessed 5.2.19).

IUPAC, 2005. Nomenclature of Inorganic Chemistry: IUPAC Recomendations, Pure and Applied Chemistry. RSC Publishing.

Izatt, R.M., Izatt, S.R., Bruening, R.L., Izatt, N.E., Moyer, B.A., 2014. Challenges to achievement

of metal sustainability in our high-tech society. Chem. Soc. Rev. 43, 2451–2475.

Jakobsson, L.K., Kennedy, M.W., Aune, R.E., Tranell, G., 2016. Recovery of rare earth elements from the ferrous fraction of electronic waste, in: Kirchan, R. et al. (Ed.), REWAS 2016: Towards Materials Resource Sustainability. Springer, pp. 89–93.

Jenkins, H., McKeag, A., Ranby, P., 1949. Alkaline earth halophosphate phosphors [WWW Document]. URL http://www.freepatentsonline.com/2488733.pdf (accessed 5.2.19).

King, A.H., Eggert, R.G., Gschneidner, K.A., 2016. Handbook on the physics and chemistry of rare earths. Volume 50, Including actinides. Elsevier B.V., North Holland.

Kourtelesis, M., Panagiotopoulou, P., Ladas, S., Verykios, X.E., 2015. Influence of the Support on the Reaction Network of Ethanol Steam Reforming at Low Temperatures Over Pt Catalysts. Top. Catal. 58, 1202–1217.

Krishnamurthy, N., Gupta, C.K., 2015. Extractive metallurgy of rare earths, Extractive Metallurgy of Rare Earths, Second Edition. CRC Press.

Kumar, L., Jain, S., 2014. Electric propulsion system for electric vehicular technology: A review. Renew. Sustain. Energy Rev. 29, 924–940.

Lakshmanan, A., 2008. Luminescence and display phosphors: Phenomena and applications, Luminescence and Display Phosphors: Phenomena and Applications. Nova Science Publishers.

Liguras, D.K., Kondarides, D.I., Verykios, X.E., 2003. Production of hydrogen for fuel cells by steam reforming of ethanol over supported noble metal catalysts. Appl. Catal. B Environ. 43, 345–354.

Liu, H., Zhang, S., Pan, D., Tian, J., Yang, M., Wu, M., Volinsky, A.A., 2014. Rare earth elements recycling from waste phosphor by dual hydrochloric acid dissolution. J. Hazard. Mater. 98, 96–101.

Liu, Z., Senanayake, S.D., Rodriguez, J.A., 2016. Elucidating the interaction between Ni and CeOx in ethanol steam reforming catalysts: A perspective of recent studies over model and powder systems. Appl. Catal. B Environ. 197, 184–197.

Mao, Z.Y., Wang, F., Chen, J.J., Wang, D.J., 2015. Rare-Earth Free Phosphors with Ultra-Broad Band Emission Application for High Color Rendering Index Lighting Source. J. Am. Ceram. Soc. 98, 3856–3862.

Meshram, P., Pandey, B.D., Mankhand, T.R., 2016. Process optimization and kinetics for leaching of rare earth metals from the spent Ni-metal hydride batteries. Waste Manag. 51, 196–203.

Mishra, B., 2015. Global Sustainability of Rare Earth Metals and Compounds, in: Engineering Solutions for Sustainability. Springer International Publishing, Cham.

Morimoto, S., Ooi, S., Inoue, Y., Sanada, M., 2014. Experimental evaluation of a rare-earth-free PMASynRM with ferrite magnets for automotive applications. IEEE Trans. Ind. Electron. 61, 5749–5756.

Müller, T., Friedrich, B., 2006. Development of a recycling process for nickel-metal hydride batteries. J. Power Sources 158, 1498–1509.

Nassar, N.T., Du, X., Graedel, T.E., 2015. Criticality of the Rare Earth Elements. J. Ind. Ecol. 19, 1044–1054.

Nguyen, L.P., Pham, Y.T.H., Ngo, P.T., Tran, T. Van, Tran, L.V., Le, N.T.H., Nguyen, L.H., Dang, T.T., Nguyen, D.A., Wenzel, M., Hartmann, D., Gloe, K., Weigand, J.J., Kretschmer, K., 2018. Production of high purity rare earth mixture from iron-rich spent fluid catalytic cracking (FCC) catalyst using acid leaching and two-step solvent extraction process. Korean J. Chem. Eng. 35, 1195–1202.

Ogi, T., Kaihatsu, Y., Iskandar, F., Wang, W.-N., Okuyama, K., 2008. Facile Synthesis of New Full-Color-Emitting BCNO Phosphors with High Quantum Efficiency. Adv. Mater. 20, 3235–3238.

Panayotova, M., Panayotov, V., 2012. Review of methods for the rare earth metals recycling. Annu.

Univ. Min. Geol. St. Ivan Rilski, Part II, Min. Miner. Process. 55, 142–147.

Parkinson, G., 1987. Reyclers try new ways to process spent catalysts. Chem. Eng. J. 94, 25–31.

Pawade, V.B., Swart, H.C., Dhoble, S.J., 2015. Review of rare earth activated blue emission phosphors prepared by combustion synthesis. Renew. Sustain. Energy Rev. 52, 596–612.

Pohl, W.L., 2011. Economic Geology Principles and Practice. Wiley-Blackwell.

Polikarpov, E., Catalini, D., Padmaperuma, A., Das, P., Lemmon, T., Arey, B., Fernandez, C.A., 2015. A high efficiency rare earth-free orange emitting phosphor. Opt. Mater. (Amst). 46, 614–618.

Reck, B.K., Graedel, T.E., 2012. Challenges in metal recycling. Science. 337, 690–695.

Reed, D.W., Fujita, Y., Daubaras, D.L., Jiao, Y., Thompson, V.S., 2016. Bioleaching of rare earth elements from waste phosphors and cracking catalysts. Hydrometallurgy 166, 34–40.

Riba, J.R., López-Torres, C., Romeral, L., Garcia, A., 2016. Rare-earth-free propulsion motors for electric vehicles: A technology review. Renew. Sustain. Energy Rev. 57, 367–379.

Roskill, 2016. Rare Earths: Global Industry, Markets and Outlook to 2026 2016. Roskill Informantion Service Ltd.

Saitoh, N., Nomura, T., Konishi, Y., 2017. Biotechnological recovery of platinum group metals from leachates of spent automotive catalysts, in: Kim, H., Alam, S., Neelameggham, N., Oosterhof, H., Ouchi, T., Guan, X. (Eds.), Rare Metal Technology. Minerals, Metals and Materials Series. Springer, Cham.

Sarapää, O., Al Ani, T., Lahti, S.I., Lauri, L.S., Sarala, P., Torppa, A., Kontinen, A., 2013. Rare earth exploration potential in Finland. J. Geochemical Explor. 133, 25–41.

Schmitt, R.A., Mosen, A.W., Suffredini, C.S., Lasch, J.E., Sharp, R.A., Olehy, D.A., 1960. Abundances of the rare-earth elements, lanthanum to lutetium, in chondritic meteorites. Nature 186, 863–866.

Schüler, D., Buchert, M., Liu, R., Dittrich, S., Merz, C., 2011. Study on Rare Earths and Their Recycling, Öko-Institut.

Shinde, K.N., Dhoble, S.J., Swart, H.C., Park, K., 2012. Phosphate Phosphors for Solid-State Lighting. Springer.

Smith, B.J., Eggert, R.G., 2016. Multifaceted Material Substitution: The Case of NdFeB Magnets, 2010–2015. JOM 68, 1964–1971.

Smith Stegen, K., 2015. Heavy rare earths, permanent magnets, and renewable energies: An imminent crisis. Energy Policy 79, 1–8.

Solvay, 2012. Solvay launches its rare earth recycling activity in France: Investment worth more than fifteen million euros to recycle rare earths from the "urban mine" [WWW Document]. URL https://www.investegate.co.uk/solvay-s-a-/gnw/solvay-launches-its-rare-earth--recycling-activ---/20120927110614H3970/ (accessed 5.2.19).

Sonoc, A., Jeswiet, J., Soo, V.K., 2015. Opportunities to improve recycling of automotive lithium ion batteries, in: Procedia CIRP.

Steinlechner, S., Antrekowitsch, J., 2015. Potential of a Hydrometallurgical Recycling Process for Catalysts to Cover the Demand for Critical Metals, Like PGMs and Cerium. JOM 67, 406–411.

Suppo, 2006. NiMH Battery vs NiCd Battery [WWW Document]. URL http://www.suppo.com/v1/NiMH vs NiCd and Li-ion battery_2014916531.pdf (accessed 5.2.19).

Svoboda, J., 2004. Magnetic techniques for the treatment of materials. Kluwer Academic Publishers.

Trovarelli, A., 2002. Catalysis by Ceria and Related Materials, Catalytic Science Series. Imperial College Press.

Tunsu, C., Ekberg, C., Retegan, T., 2014. Characterization and leaching of real fluorescent lamp waste for the recovery of rare earth metals and mercury. Hydrometallurgy 144–145, 91–98.

Tunsu, C., Petranikova, M., Gergorić, M., Ekberg, C., Retegan, T., 2015. Reclaiming rare earth elements from end-of-life products: A review of the perspectives for urban mining using hydrometallurgical unit operations. Hydrometallurgy 156, 239–258.

U.S. Department of Energy, 2011. Critical Materials Strategy [WWW Document]. URL https://www.energy.gov/sites/prod/files/DOE_CMS2011_FINAL_Full.pdf (accessed 5.2.19).

U.S. Department Of Energy, 2015. Quadrennial Technology Review: An Assessment of Energy Technologies and Research Opportunities [WWW Document]. Quadrenn. Technol. Rev. URL https://www.energy.gov/sites/prod/files/2017/03/f34/quadrennial-technology-review-2015_1.pdf

Umicore, 2018. Umicore - Our recycling process [WWW Document]. URL https://csm.umicore.com/en/recycling/battery-recycling/our-recycling-process (accessed 5.2.19).

United Nation, 1987. Report of the World Commission on Environment and Development: Our Common Future [WWW Document]. Sustain. Dev. URL https://sustainabledevelopment.un.org/content/documents/5987our-common-future.pdf

Van Loy, S., Binnemans, K., Van Gerven, T., 2017. Recycling of rare earths from lamp phosphor waste: Enhanced dissolution of LaPO4:Ce3+,Tb3+ by mechanical activation. J. Clean. Prod. 156, 226–234.

Walton, A., Yi, H., Rowson, N.A., Speight, J.D., Mann, V.S.J., Sheridan, R.S., Bradshaw, A., Harris, I.R., Williams, A.J., 2015. The use of hydrogen to separate and recycle neodymium-iron-boron-type magnets from electronic waste. J. Clean. Prod. 104, 236–241.

Wang, J., Tatsumoto, M., Li, X., Premo, W.R., Chao, E.C.T., 1994. A precise 232Th-208Pb chronology of fine-grained monazite: Age of the Bayan Obo REE-Fe-Nb ore deposit, China. Geochim. Cosmochim. Acta 58, 3155–3169.

Wang, W.-N., Ogi, T., Kaihatsu, Y., Iskandar, F., Okuyama, K., 2011. Novel rare-earth-free tunable-color-emitting BCNO phosphors. J. Mater. Chem. 21, 5183–5189.

Widmer, J.D., Martin, R., Kimiabeigi, M., 2015. Electric vehicle traction motors without rare earth magnets. Sustain. Mater. Technol. 3, 7–13.

Wu, Y., Wang, B., Zhang, Q., Li, R., Sun, C., Wang, W., 2014. Recovery of rare earth elements from waste fluorescent phosphors: Na2O2 molten salt decomposition. J. Mater. Cycles Waste Manag. 16, 635–641.

Xu, G., Yano, J., Sakai, S. ichi, 2016. Scenario analysis for recovery of rare earth elements from end-of-life vehicles. J. Mater. Cycles Waste Manag. 18, 469–482.

Yang, F., Kubota, F., Baba, Y., Kamiya, N., Goto, M., 2013. Selective extraction and recovery of rare earth metals from phosphor powders in waste fluorescent lamps using an ionic liquid system. J. Hazard. Mater. 254–255, 79–88.

Yang, X., Zhang, J., Fang, X., 2014. Rare earth element recycling from waste nickel-metal hydride batteries. J. Hazard. Mater. 279, 384–388.

Yang, X., Zhao, D., Leck, K.S., Tan, S.T., Tang, Y.X., Zhao, J., Demir, H.V., Sun, X.W., 2012. Full Visible Range Covering InP/ZnS Nanocrystals with High Photometric Performance and Their Application to White Quantum Dot Light-Emitting Diodes. Adv. Mater. 24, 4180–4185. https://doi.org/10.1002/adma.201104990

Yang, Y., Walton, A., Sheridan, R., Güth, K., Gauß, R., Gutfleisch, O., Buchert, M., Steenari, B.-M., Van Gerven, T., Jones, P.T., Binnemans, K., 2016. REE Recovery from End-of-Life NdFeB Permanent Magnet Scrap: A Critical Review. J. Sustain. Metall. 3, 122–149.

Zakotnik, M., Harris, I.R., Williams, A.J., 2009. Multiple recycling of NdFeB-type sintered magnets. J. Alloys Compd. 469, 314–321.

Zeng, X., Li, J., Singh, N., 2014. Recycling of spent lithium-ion battery: A critical review. Crit. Rev. Environ. Sci. Technol. 44, 1129–1165.

Zhan, F., Jiang, L.J., Wu, B.R., Xia, Z.H., Wei, X.Y., Qin, G.R., 1999. Characteristics of Ni/MH

power batteries and its application to electric vehicles. J. Alloys Compd. 293–295, 804–808.

Zhan, W., Guo, Yun, Gong, X., Guo, Yanglong, Wang, Y., Lu, G., 2014. Current status and perspectives of rare earth catalytic materials and catalysis. Cuihua Xuebao/Chinese J. Catal. 35, 1238–1250.

Zhao, Z., Qiu, Z., Yang, J., Lu, S., Cao, L., Zhang, W., Xu, Y., 2017. Recovery of rare earth elements from spent fluid catalytic cracking catalysts using leaching and solvent extraction techniques. Hydrometallurgy 167, 183–188.

Zheng, T., He, Junjun, Zhao, Y., Xia, W., He, Jieli, 2014. Precious metal-support interaction in automotive exhaust catalysts. J. Rare Earths 32, 97–107.