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Addressing Criticality in Rare Earth Elements via Permanent Magnets Recycling

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Abstract

Rare earth elements (REEs) are critical for many advanced technologies and are faced with potential supply disruptions. Recycling of permanent magnets (PMs) can be good sources for REEs which can help minimize global dependence on freshly mined REEs, but PMs are rarely recycled. Recycling of PMs has been discussed with respect to improving REEs resource sustainability. Some challenges to be addressed in order to establish industrially deployable technologies for PMs recycling have also been discussed, including profitability, energy efficiency and environmental impacts. Key considerations for promoting circular economy via PMs recycling is proposed with the focus on deciding the target points in the supply chain at which the recycled products will be inserted. Important technical considerations for recycling different forms of waste PMs, including swarfs, slags, shredded and intact hard disk drives magnets, have been presented. The aspects of circular economy considered include reusing magnets, remanufacturing magnets and recovering of REEs from waste PMs.

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ABSTRACT

Rare earth elements (REEs) are critical for many advanced technologies and are faced with potential supply disruptions. Recycling of permanent magnets (PMs) can be good sources for REEs which can help minimize global dependence on freshly mined REEs, but PMs are rarely recycled. Recycling of PMs has been discussed with respect to improving REEs resource sustainability. Some challenges to be addressed in order to establish industrially deployable technologies for PMs recycling have also been discussed including profitability, energy efficiency and environmental impacts. Key considerations for promoting circular economy via PMs recycling is proposed with focus on deciding the target points in the supply chain at which the recycled products will be inserted. Important technical considerations for recycling different forms of waste PMs including swarfs, slags, shredded and intact hard disk drives (HDDs) magnets, were presented. The aspects of circular economy considered include reusing magnets, remanufacturing magnets and recovering of REEs from waste PMs.

INTRODUCTION

A technology designed for circular economy should reduce materials and energy wastes, promote robust product designs, encourages reuse, remanufacturing and recycling of resources. Recycling of rare earth elements (REEs) permanent magnets (PMs) can be an important aspect of circular economy and sustainability in REEs resource. In the context of this article, recycling includes reusing PMs, reprocessing/remanufacturing PMs and recovering REEs from PMs.

REEs are essential for the modern economy because they enable many technologies; computing, lighting, automation, transportation, national defense systems and many clean energy applications. In many of these applications, only small amounts of REEs are required but significant amounts of some REEs are used in permanent magnet alloys. The 4*f* electrons in REEs play important role in generating high magnetocrystalline anisotropy, which is an intrinsic magnetic property and a figure of merit indicating the suitability of materials for developing PMs.

Current commercial REE PMs are based on neodymium-iron-boron (Nd-Fe-B), samarium-cobalt (Sm-Co) and samarium-iron-nitrogen (Sm-Fe-N) compounds. Praseodymium (Pr), co-mined with Nd, is often found in Nd-Fe-B magnets largely because of the difficulty and cost of separating both elements. Dysprosium and terbium are often added to Nd-Fe-B PMs to enhance coercivity and improve high temperature performance. Transition metals such as cobalt, gallium and copper are also added to improve PMs processing and magnetic properties. Sm_2Co_{17} also contain iron, copper and zirconium while $SmCo_5$ can contain gadolinium and terbium. Although promising on the basis of its magnetic performance indicators, the inability to consolidate Sm-Fe-N by conventional sintering process has limited its applications [1].

REE PMs are resources that are being used at an accelerating rate. Emerging technologies that significantly depend on the same REEs can disrupt the demand-supply balance and result in market uncertainties and associated price spikes or constraints on technology deployment. Growth of existing technologies such as electric vehicles and permanent magnet wind generators is also capable of triggering such price spikes. The lopsided geographical distribution of REE production in favor of China has been known to trigger supply disruption and price spike due to export restrictions [2]. With no new mines being established and existing mines closing [3], it is reasonable to expect supply shortages to trigger price spikes, which may pose direct threats to technologies that depend on strong PMs.

Brief Review of Permanent Magnets Market Trends

The accelerating shift towards cleaner and more efficient technologies results in increasing electrification and the need to use more REE-based PMs in energy conversion devices. International Market Analysis Research and Consulting (IMARC) reported that during 2005–2012, the global market for REE PMs grew at a compound annual growth rate (CARG) of ~9% [4]. In 2013, over 106,000 metric tons of REE containing magnets, worth over $11 billion were sold [4]. These magnets were used in motors, generators, medical equipment, consumer and appliances, speakers and other applications. Motors and generators account for >30% of REE-based PMs sales [4–6]. Also during 2010–2015, the amount of rare earth oxides (REOs) used for magnet production increased from 21%–26% [7]. A 9.4% CARG has been estimated for only Nd-Fe-B during 2013–2020 [8]. A 2017 forecast estimated a CARG of 8.6% and markets value of $12.3 billion by 2022 [6].

During 2005–2013, nearly 97% by volume of the REE PMs sold was based on Nd-Fe-B. In 2008, about 21% of all REEs production was used for Nd-Fe-B magnets [9,10]. In 2016, the market for Nd-Fe-B magnet was valued at ~$7.5 million [6]. It is projected that by 2019 the global demand for Nd-Fe-B and Sm-Co magnets will reach 173,500 and 4000 metric tons, respectively [4]. A 2016 market research forecast shows that by 2020, 137,200 and 5,400 metric tons of Nd-Fe-B and Sm-Co PMs, respectively, will be used in applications [5]. If 30% [11] of the original materials were lost in PMs manufacturing process, by 2020 Nd-Fe-B and Sm-Co production can reach 196,000 and ~7,700 metric tons, respectively.

RECYCLING PERMANENT MAGNETS: A SECONDARY SOURCE FOR REEs SUPPLIES

Recycling presents a viable strategy to help mitigate the supply risks associate with REEs. It can help improve resource sustainability by re-introducing REEs into the supply chain and reducing the burden on freshly mined ores. Recycling is well established in many aspects of society: the Institute of Scrap Recycling Industries has estimated that over 40% of industrial consumers' raw materials come from recycling [12]. Recycling REE-containing materials can enable production from secondary raw materials and help recover the embodied energy in used materials. If well applied, REEs recycling should have significantly less negative impact on the environment than obtaining materials from primary sources [13]. It will also eliminate exposures to radioactive elements, typical of mining REEs from ores.

Availability of Permanent Magnets Recycling Feedstock

Insufficient recycling feedstock is often cited as a barrier to REEs recycling. However, the increasing use of REEs in applications suggests that insufficient feedstock is not yet a barrier since significant REE-containing waste materials are currently not being recycled. Nevertheless, as more REEs are recycled, insufficient recycling feedstock may become a barrier. Currently, difficulty in accessing and collecting REE-containing materials and devices may be the most significant challenge [14]. This challenge can be addressed by focusing on applications, such as PMs, in which recycling feedstock materials are concentrated at places where they can be easily accessed. Apart from catalysis, which typically uses low-value cerium-based materials, PMs consume the most significant amount of global REEs production including the high-value elements; Dy, Pr, Nd and Sm. The consumption of these elements will increase with the increasing market-share of clean energy technologies [15].

Magnet manufacturing and processing plants can be sources of significant amounts of magnets for recycling. It is estimated that 30% or more of an original material for magnet manufacturing can be generated as waste in such plants [11]. Such waste PMs are generated as swarfs and slags with varying degrees of oxidation and contaminations. Also accidentally damaged PMs during manufacturing or post-manufacturing processing can be recycled. Electronic devices, such as hard disk drives (HDDs) can be collected for recycling. HDDs are particularly concentrated in data centers. For example, 557 million HDDs were shipped in 2014 [16]. A recent study reported that REEs from U.S. HDDs alone could meet ~5.2% of global (excluding China) Nd-Fe-B magnets demand [14]. Good progress has recently been in developing technologies for extracting magnets from HDDs. In December 2010, Hitachi developed machinery for separating and collecting, and process for extracting PMs from end-of-life systems [17]. The Critical Materials Institute recently developed an automated conveyor-type line system for extracting PMs from one HDD per four seconds [18,19]. The University of Birmingham also reported the use of hydrogen gas to extract magnets from HDDs [20–22].

It is important to understand how much REEs can potentially be obtained from recycling PMs. Fig. 1 shows volumes of Nd-Fe-B and Sm-Co, by application area, used in 2014, 2015 and forecasted for 2020 [5]. Electric motors and consumer electronics are forecasted to continue to dominate the use of REE PMs. Swarfs generated during manufacturing of PMs for those applications should be readily available for recycling. Comparing Fig. 1 to the lifespan of devices in the applications, additional PMs available for recycling can be inferred. As an example, assuming six years lifespan for consumer electronics [14], ~36,000 metric tons of Nd-Fe-B and ~1,600 metric tons of Sm-Co produced in 2020 could be available for recycling by 2026. The volumes may be higher, depending on how much the consumer electronics made before 2020 were recycled. On the other hand, motors typically have longer lifespans (10–20 years) [23,24]. As a result, REEs from PMs in motors would take longer to become available for recycling. The same applies to medical devices and wind turbines which typically have long average lifetimes [25].

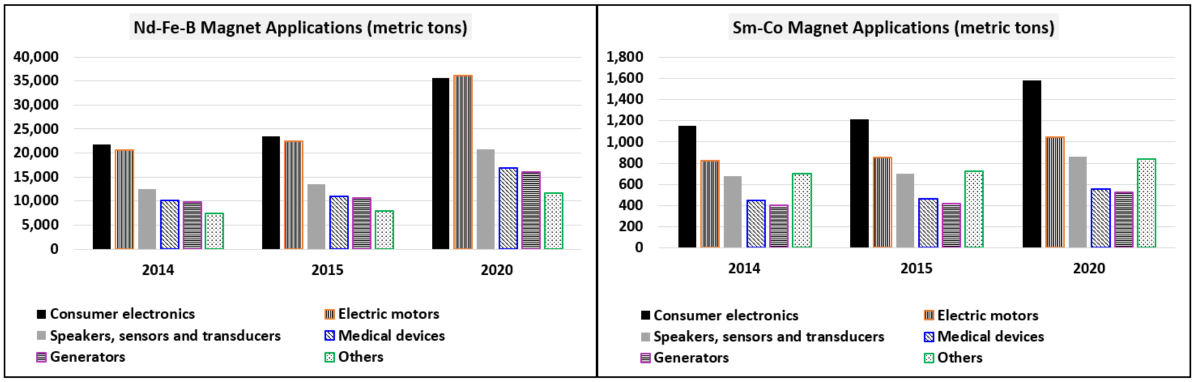

Fig. 1: 2014, 2015 and 2020 (forecasted) applications of Nd-Fe-B and Sm-Co PMs (*plotted with data from* [5]).

CHALLENGES TO RECYCLING OF REEs IN PERMANENT MAGNETS

Challenges in Justifying Recycling

Despite many proposed technologies for recovering REEs from PMs, most are not commercially deployed which limits the resource sustainability potential for recovering REEs from PMs. A bottleneck to recycling REEs from PMs is the cyclical nature of the instability in REEs prices. Response by governments and other institutions when price spikes occur help drive the need to recycle [26]. The 2011 price spike led to the establishment of efforts across the world including the U.S. Department of Energy Critical Materials Institute [27], the European Innovation Partnership on Raw Materials [28] and other efforts in different regions of the world. It also resulted in international coordinated efforts to develop strategies for mitigating supply disruptions [29]. Justifying that recycling can help mitigate supply disruptions is less challenging during price spikes but the strength of such argument wanes as prices decrease again; making recycling appear less feasible. In Fig. 2 [30,31], it would be easier to justify establishing a recycling facility in 2011 compared to 2010 or 2016. Nevertheless, it is advantageous to have readily deployable REEs recycling technologies, should high prices return, and persist.

Profitability Challenges

Another challenge to recycling REEs is that high capital expenditure (CAPEX) and operating expenditure (OPEX) can make establishing some of the proposed processes unprofitable. Taking HDDs for example, we obtained an average magnet weight of 13.3g per HDD from 25 HDDs, which lies between 10g per HDD [32,33] and 16.3g per HDD [34] reported by others. Stoichiometric $Nd_2Fe_{14}B$ contains ~27% REEs by weight, although 24.4 – 31%, averaging 28.2%

was reported from x-ray fluorescence studies performed using 10 HDDs [34]. Therefore assuming 13.3g of magnet per HDD and 28.2 wt.% REE per magnet, an average magnet in one HDD would contain ~3.8g of REEs. Establishing a profitable recycling enterprise based on such small amount of REEs requires low OPEX and the CAPEX needs be amortized over a short period to protect the enterprise from the potential impacts of falling prices. This is particularly challenging when HDDs are shredded for security reason because additional costs would be needed to collect, pre-sort, demagnetize and concentrate the PMs content of the shredded HDDs, before REEs recovering can be performed. This challenge is further exacerbated in HDDs with smaller magnets.

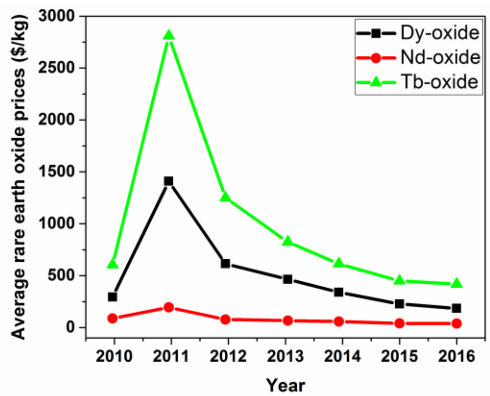

Fig. 2: 2010–2016 trends in Dy-oxide, Nd-oxide and Tb-oxidw prices: comparing the ease of justifying REEs recycling based on prices (*plotted with data* [30,31])

Three possible approaches for improving the profitability of recovering REEs are employing processes that: (a) allow for recycling of other components in a system containing REEs (b) result in valuable recycling by-products and (c) eliminate/minimize pre-processing steps prior to recovering REEs. A process reported by the Critical Materials Institute may have these features [35]. The details of the process are yet to be made public due to the need to protect intellectual properties.

Safety, Environmental Impact and Energy Efficiency Concerns

REE recycling efforts also need to be safe to operate, environmentally friendly and energy efficient. For operational safety, limiting or avoiding the generation of hazardous fumes is necessary. For example, processes that result in the evolution of harmful gasses (e.g. hydrogen gas [36] as in the use of β-diketonates) may be difficult to scale up, in addition to posing some safety concerns. Also processes that use large amounts of chemicals will raise environmental concerns, especially ones that generate non-recyclable by-products with no economic values. This environmental concern is one strong drawback of most hydrometallurgical processes, which often use large amount of chemicals and generate large amount of wastewaters [37]. Moreover, hydrometallurgical processes often depend on strong mineral acids to dissolve the magnets [38–43]. Since acid digestion of magnets is typically non-selective, large amount of acid-contaminated wastes are typically generated. Although gas-phase extraction [37] and other pyrometallurgical processes do not generate large amounts of contaminated wastewaters, they require highly corrosive gases, use large amounts of energy and generate large amounts of solid wastes [37]. Additional investments will be required to overcome these safety and environmental challenges. The consequence is higher operating expenditure and reduced revenue.

Although it is estimated that recovering REEs from PMs can be more energy efficient and environmental friendly than obtaining from primary sources [13], it is still important to minimize the energy used and impact on the environment. This can be appreciated by considering that the initial production process for REOs used in manufacturing PMs is energy intensive and environmentally impactful. Fig. 3 shows the environmental footprint associated with the production of the REOs mostly used in manufacturing PMs [44]. Putting the energy numbers in perspective, the combined energy for production of only 1 kg of these oxides (Nd-oxide, Pr-oxide, Sm-oxide and Dy-oxide) is more than the average monthly energy consumed in the U.S. State of Texas in 2016 (1156 kWh = 4161 MJ [45]). Reducing the REOs into metals and subsequent manufacturing of PMs also add to the total energy and greenhouse gas emission.

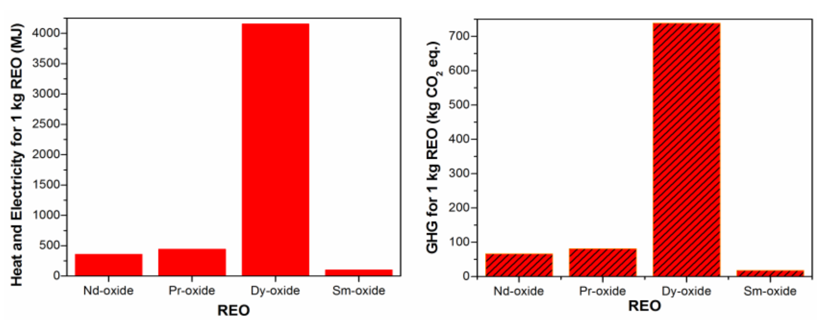

Fig. 3: Energy and greenhouse gas emission associated with the production of 1 kg of primary REOs: Nd_2O_3, Pr_6O_{11}, Sm_2O_3 and Dy_2O_3, for magnet production. (*plotted with data from* [44]).

CONSIDERATIONS FOR REEs PERMANENT MAGNETS RECYCLING

Recycling PMs requires deciding whether to directly reuse the PMs or recover the REEs contents. This decision depends on the point in the supply chain at which the product of recycling will be inserted. Fig. 4 is a decision tree for recycling PMs, which also demonstrates the potential for circular economy. The figure considers: (a) need to recycle other materials housed in the same system as the PMs (b) assessment of the PMs to determine suitability for reuse or need for reprocessing before reuse (c) requalification of remade PMs for intended applications, and (d) recovering of REEs when reuse is not possible or would result in expensive reprocessing. For reuse, minimal or no reprocessing of the PMs is ideal. An example is the magnet-to-magnet recycling approach employed by Urbanmining Company [46]. Recovering the REEs contents as metals, oxides or other compounds enables wider application of the recycled products as raw materials for making new products. However, additional investments in energy and other resources may be needed. In general, the decision to reuse or recover the REEs should minimize negative environmental impacts and the resource investments needed to obtain a useful products. Recycling other components of the system in which the permanent magnet was contained can help in making a recycling process more profitable.

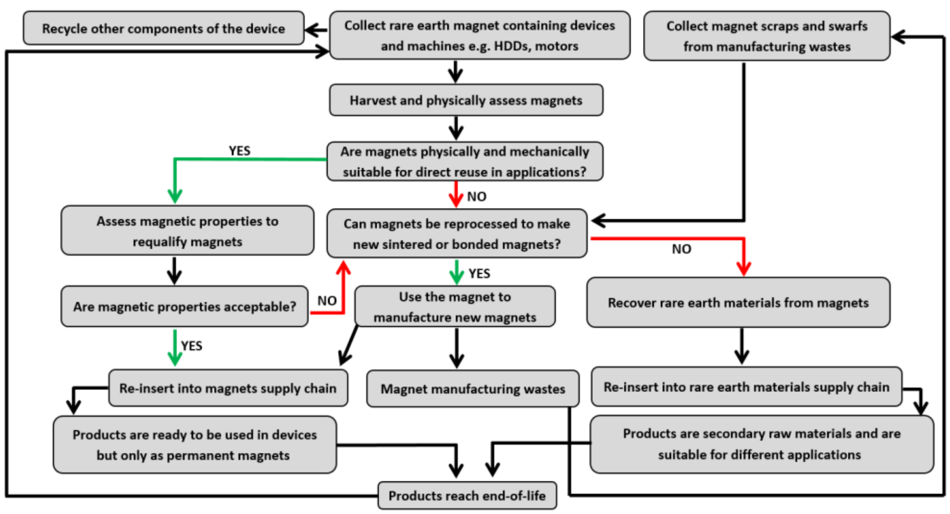

Fig. 4: Decision diagram illustrating circular economy for recycling REEs from permanent magnets materials contained in devices and magnet manufacturing plants.

PERMANENT MAGNET MATERIALS FOR RECYCLING

Reusing Permanent Magnets

Waste PMs for recycling can be swarfs or slags, which can be dry or wet. Fig. 5A and 5B show pictures of industrially obtained dry and wet magnet swarfs, respectively. Swarfs are fine particles of PMs obtained during grinding, cutting and other post-manufacturing operations. Since Sm-Co has better oxidation resistance than Nd-Fe-B, swarfs and of Sm-Co are typically less oxidized. Swarfs are also typically contaminated with cutting/grinding fluid and media. Slags, such as shown in Fig. 5C, are typically unsuitable for reuse.

Fig. 5: Forms of feedstock materials for PMs recycling: (A). Dry grinding swarfs (B). Wet grinding swarfs (C). Slags (D). Shredded electronic waster from HDD (E). Scrap magnets and (F) HDD magnets.

Fig. 5D shows PMs-containing shredded HDD. Although done for data security, shredding HDDs adds more difficulty to PMs recycling, especially for reusing the PMs content. Separating the PMs (typically Nd-Fe-B) in as-shredded HDDs from other ferrous (e.g. Ni) and non-ferrous components can be challenging, which makes it difficult to reuse PMs from shredded HDDs. Shredding also results in the loss of PMs, which reduces the amount of recyclable materials [47,48].

Figs. 5E and 5F show scrap magnets from industrial wastes and magnets extracted from HDDs using an aforementioned process [18]. Ideally both types of materials can be reprocessed to remake sintered magnets [20,49,50], produce isotropic and anisotropic bonded PMs [51–54] or recover the REEs from the PMs.

Shredded HDDs (Fig. 6A) with high concentration of PMs can be demagnetized (Fig.6B) and the PMs content separated (Fig. 6C). In Fig. 6D, the magnetic hysteresis plot for the as-shredded material (black plot) is typical of decoupled soft and hard magnetic phases; nickel and Nd-Fe-B, respectively. The plot for the separated PMs (red plot) is typical of Nd-Fe-B magnet. Fig. 6D and the inset table show that, if well separated, the PMs content of shredded HDDs can be reused. This obviously depends on the form of materials obtained from the shredding process: coarse or fine.

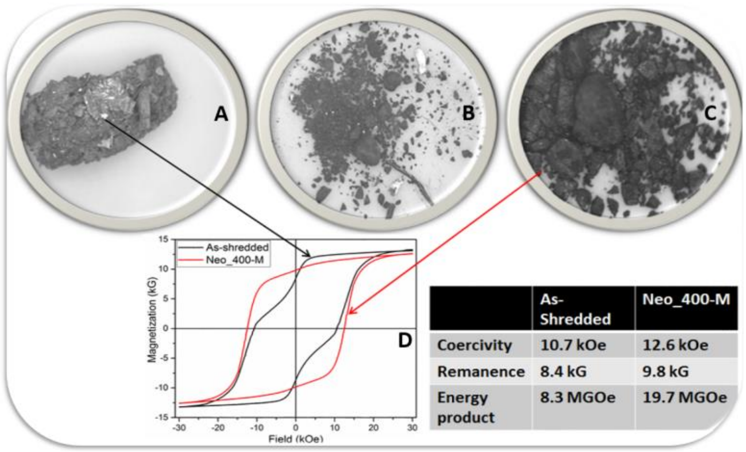

Fig. 6: Demonstrating the possibility to reuse magnets from shredded HDDs showing: (A) Magnet manually concentrated from as-shredded HDD; (B) Material derived from demagnetizing the magnet concentrate; (C) Magnet separated from other components of the mix; (D) Magnetic hysteresis loops of the magnet concentrate (black plot) and the separated magnet (red plot). Inset: Magnetic properties derived from the hysteresis loops.

Recovering REEs Contents of Permanent Magnets

When reuse is not possible, the REEs content of the materials can be recovered. Others have discussed the different methods for recovering the REEs, including the advantages and disadvantages of the methods[37,55]. In general, REEs are recovered as oxides (typical of many hydrometallurgical approaches) or metals (typical of some pyrometallurgical approaches).

Abrahami et al.[56] have investigated recovery of REEs from post-consumer shredded HDDs by sulfuric acid leaching and by combining molten slag extraction process and sulfuric acid leaching. The recycling feedstock materials for both routes were concentrated PMs obtain after subjecting as-shredded HDD to thermal demagnetization, grinding and screening. Both routes resulted in 98.4% pure REE double salt from which REE-fluorides or oxides can be obtained.

The lack of a mature technology for recovering REEs from end-of-life (EOL) electronic devices [57] appears to be related to the challenges with obtaining REEs from EOL devices. As previously stated, shredding of HDDs complicates this problem by resulting in pre-processing steps before any of the known routes for recovering REEs can be applied. The complications can be overcome by developing a process that selectively recovers the REEs without the need for pre-processing the shredded HDDs (manual picking and sorting, demagnetization, grinding and sieving). Such process will reduce the complexity of recycling processes, help avoid co-melting or co-dissolution of unwanted device components, and eliminate additional investments for generating and containing wastes. As previously stated, selective recovery of REEs from electronic wastes in the as-shredded form is a features of the new technologies announced by the Critical Materials Institute [35].

When recovery of REEs is the option, different routes have been developed. Carlson and Taylor reported on selective sulfation roasting process in which the difference in the stabilities of sulfates of REEs and transition metals are exploited to separate the REEs from Nd-Fe-B [58]. Stanton reported on separation of Sm from Sm-Co, also using the selective sulfation approach [59]. The efficiency of different types of acid for REEs leaching has been studied, in which hydrochloric and sulfuric acids were found most effective for complete dissolution of the magnets [60]. Recovery of REEs from both Nd-Fe-B and Sm-Co recycling feedstock via ionic liquid routes has also been proposed [61–65]. One challenge with the ionic liquid routes is that ionic liquids are currently expensive compared with many of other solvents therefore limiting the potential for large scale recycling [66]. Hydrothermal process was reported to result in selective recovery of Nd compound with >99% purity from Ni-coated magnet. However the authors remarked that large amount of acidic waste water was generated and needs to be cleaned up [67]. Pyrometallurgical approaches have been studied and developed for recovering both light rare earth elements and heavy rare earth elements from magnet scraps via the liquid-metal extraction process [68–71] and electroslag refining methods [38,72].

CONCLUSION

REE-based PMs have been and will continue to be of critical importance for the advancement of modern technologies. Although there is currently scarcity of well-established and industrially adopted REEs recycling technology for PMs, there can be sufficient material to encourage recycling, irrespective of the waning in the motivation to recycle when REEs prices decline. Those materials for recycling include existing materials and those that will arise from increased use of PMs in the future. The current decline in REEs prices is far removed from being a security for future supplies. As a result, establishment of recycling processes to be deployed, when prices spike, is very important.

The challenges with establishing industrial recycling practices is likely a consequence of the lack of deployable recycling technologies. Many of the proposed approaches have drawbacks which can deter new start-up efforts and discourage existing companies from integrating the process into their plants. Those drawbacks, including the use and generation of hazardous fumes and chemicals, consumption of large amounts of energy need to be addressed to encourage adoption of the processes. Simplifying complex process can also be very useful.

Since businesses are profit-driven, proposed technologies for recycling REEs from PMs must demonstrate profitability to encourage adoption by the private sector. Preferably, process should minimize waste generation or result in valuable by-products for use in the same or other processes. Profitability should be considered from system perspective, rather than single material perspective; although process costs, energy requirements and environmental impacts also need to be considered from the same system perspectives.

Part of the decision to recycle should be the consideration to reuse, where the REE PMs can be qualified as fit for an application. When direct reuse is not possible, the material can be considered for applications for which minimal processing is possible for remanufacturing of PMs. Recovery of the REEs, as metals or oxides, should be considered only when the materials cannot be directly reused or minimally processed to remanufacture PMs.

Finally, considering several reports of breakthroughs by researchers, national and international coordinated strategies and consortiums on recycling, it seems very likely that new REE recycling efforts capable of addressing many of the concerns will be soon established.

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