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Recycling of the rare earth elements

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The rare earth elements (REE) are vital to modern technologies and society and are amongst the most critical of the critical elements. Despite these facts, typically only around 1% of the REE are recycled from end-products, with the rest deporting to waste and being removed from the materials cycle. This paper provides an overview of the current and future potential of the recycling of the REE, including outlining the significant but currently unrealised potential for increased amounts of REE recycling from end-uses such as permanent magnets, fluorescent lamps, batteries, and catalysts. This future potential will require a significant amount of research but increasing the amount of REE recycling will contribute to the overcoming some of the criticality issues with these elements. These include increased demand, issues over security of supply, and overcoming the balance problem where primary mine-derived sources overproduces lower demand REE without necessarily meeting demands for the higher demand REE.

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Introduction

The International Union of Applied and Pure Chemistry (IUPAC) defines the rare earth elements (REE) as the 15 lanthanide elements plus Sc and Y [1]. Although the REE have similar electron configurations, they also have distinctive physical and chemical properties that enable their use in a broad range of technologies (Table 1). These properties mean that the REE provide special magnetic, luminescence and strength characteristics to the end-products they are used in (e.g., Ref. [2]). The properties of the REE are chemically derived from the fact that the majority of these elements have partially occupied 4f electron orbitals, making these elements often the only choice (i.e., no possible substitution) for uses in a wide range of advanced industrial applications (e.g., Ref. [2]). This has led to the REE being crucial to a wide range of modern technologies, including uses in magnets, batteries, glass, and alloys, all of which are critical to the manufacturing of modern computers, magnets, lasers, and screens [3–5].

Current global primary REE production is about 130,000 metric tons of rare-earth oxide (REO) equivalent content per year [6], with the 2014 REE market worth ~US$2051 million [5], although the small amount of recycling of these elements means it is currently unclear how recycling could affect the REE market. Global demand for these elements has steadily increased although prices have remained steady after a significant spike caused by the application and subsequent removal of Chinese export restrictions (e.g., [5]) and a subsequent oversupply of some of the REE. The majority of REE consumption is by mature markets (e.g., catalysts, glassmaking, lighting and metallurgy; 59% in 2011; [7]) with newer high growth markets such as magnets, ceramics and batteries taking up the remaining 41% [7]. The increase in usage of the REE has led to a coincident increase in demand for these elements (e.g., [5]), although the vast majority of demand is still met by primary production from mines, primarily within China, which dominated the global consumption of 119,000 t of rare earth oxides (REO) in 2014 [8]. However, one of the main issues with REE mine production is the so-called balance problem, where the vast majority of REE production is dominated by La and Ce but the majority of REE demand is for Nd or Dy (e.g., [9,10]). This could lead to a situation where La and Ce are overproduced, and the demand for Nd or Dy for use in end-products such as magnets or batteries may not be met by primary production alone. However, this issue can at least partly be overcome by recycling as the products that would be recycled predominantly contain the potentially undersupplied Nd or Dy rather than the potentially overproduced lighter REE such as La or Ce [11]. One of the main obstacles to the recycling of these elements is the fact that the amount of REE used in end-products ranges from <mg to several kg (e.g., Ref. [12]). This, combined with the complexity of their uses, the difficulty inherent in separating the individual REE from each other to yield pure single elements, the sometimes long life of certain uses (e.g., permanent magnets in electrical technologies) and a variety of other more general reasons (Table 2) means that <1% of the

Table 1

Summary of the rare earth elements and their common uses (adapted from Ref. [5]).

Element	Common uses
La	Optics, batteries, catalysis
Ce	Chemical applications, colouring, catalysis
Pr	Magnets, lighting, optics
Nd	Magnets, lighting, lasers, optics
Pm	Limited use due to radioactivity, used in paint and atomic batteries; very rare in nature
Sm	Magnets, lasers, masers
Eu	Lasers, colour TV, lighting, medical applications
Gd	Magnets, glassware, lasers, X-ray generation, computer applications, medical applications
Tb	Lasers, lighting
Dy	Magnets, lasers
Ho	Lasers
Er	Lasers, steelmaking
Tm	X-ray generation
Yb	Lasers, chemical industry applications
Lu	Medical applications, chemical industry applications
Sc	Alloys in aerospace engineering, lighting
Y	Lasers, superconductors, microwave filters, lighting

REE used today are recycled [13]. The recycling that does take place is generally in the form of REE within permanent magnets, fluorescent lighting, batteries, and the REE that are used as catalysts within the chemical industry, including in petroleum refining (e.g., [14]). However, advances are being made in the recycling of these critical elements and this review outlines the current status of REE recycling (also summarised in Table 3) as well as areas with potential for increased recycling of these critical elements.

Recycling of the REE

The small amount of the REE used in the majority of end-products containing these elements combined with the difficulty of the collection, extraction, and recovery of the constituent materials within end-products has hampered the recycling of the REE to date [15,16]. The recycling that does take place can be split into three types, namely the direct recycling of manufacturing scrap or residues, the urban mining of end-of-life products, and the recycling of solid and liquid industrial wastes [15]. The latter is somewhat atypical in that it utilises industrial residues rather than end-of-life resources, and as such is not considered here. An outline of the recycling potential of the REE is given in Table 4 [17], demonstrating the significant amount of the REE that could be recycled from magnets, NiMH batteries, and phosphorescent lighting alone. The material flow of the REE is also shown in Figure 1, which uses Nd in magnets as an example that illustrates current recycling as well as future potential.

Magnets

The majority of current REE recycling is from permanent magnets, but even this is in relatively low amounts via approaches that include [12]:

Table 2

Summary of the main barriers to effective recycling of e-waste, and the applicability to the REE; adapted from Ref. [2].

Barriers to e-waste recycling	Applicable to the REE?
End-products contain small amounts of metals targeted for recycling (g to <mg)	Yes
Lack of economic incentive to recycle as a result of low metal value per unit; primary sources used instead	Yes, after reduction in REE prices associated with removal of Chinese export restrictions in 2014
Current commercial recycling technologies cannot recover the small amounts of the metals in question that are present in modern products not adequate for recovering small amounts of metals present in modern products	Yes, but laboratory experiments that may scale to industry may remove this barrier
End-products to be recycled contain a complex mixture of metals that change over time as a result of technological advances	Yes, although the recycling of misch metals and REE alloys may remove some obstacles
End-product collection procedures are scarce or do not exist	Yes, although less the case for e.g., REE magnets
Prohibitive cost of the collection and transportation of end-products to recycling facilities	Unclear
The recycling process is not part of a collection chain that incorporates smelters	Unclear
End-product design and incorporation of target metals makes separation of recyclable material difficult	Yes
Public awareness of impending loss of crucial resources is low	Somewhat; the public are aware of the criticality of the REE but there are abundant REE resources already known, (e.g., Ref. [5])

- Traditional hydrometallurgical recovery techniques where magnets are dissolved in acids (or potentially in the future using ionic liquids) before the REE are precipitated out of solution.
- Pyrometallurgical recovery techniques where REE alloys are remelted, are separated from alloyed transition melts in a liquid metallic state, are refined in an electroslag process, or are dissolved out of alloys by reaction with a molten flux, with the REE then supercooling with the flux to form a glass. The approach used depends on the nature of the REE alloys within the magnets.

Table 3

Summary of the potential sources of REE during recycling.

Source for recycling	References	Targeted REE	Primary recycling mechanism
Industrial process & residues			
Fluid Catalytic Cracking (FCC) catalysts	[32,33,40–44]	LREE (La, Ce)	Hydrometallurgical processes (leaching, solvent extraction, selective precipitation); Microbial leaching (bioleaching)
Other industrial processes and residues	[29–31,40,45–54]	Depending on the source material, the REE recycling process can target different REE	Pyrometallurgical processes (roasting, calcination); Hydrometallurgical processes (leaching, solvent extraction, selective precipitation); Physical separation & microbial leaching (bioleaching)
WEEE & 'End of Life' consumer goods			
Fluorescent material (phosphor powder, fluorescent lamps, etc.)	[12,19,21,25,55–60]	La, Ce, Tb, and Y	Pyrometallurgy (roasting, calcination); Hydrometallurgy (leaching, solvent extraction, selective precipitation); Gas phase extraction
Magnets	[11,12,25,26,61–70]	Nd, Dy and the other REE	Hydrometallurgical processes (leaching, solvent extraction, selective precipitation)
Batteries	[12,22,24]	La, Ce, Pr, and Nd	Hydrometallurgical or Pyrometallurgical recovery routes

Table 4

Recycling potential of the REE. From Ref. [12].

REE application	Estimated REE stocks in 2020 (tons)	Estimated average lifetime (years)	Estimated REE old scrap in 2020 (tons)	Pessimistic scenario: recycled REE in 2020 (tons)	Optimistic scenario: recycled REE in 2020 (tons)
Magnets	300,000	15	20,000	3300	6600
Lamp Phosphors	25,000	6	4167	1333	2333
Nickel-metal-hydride batteries	50,000	10	5000	1000	1750
Total	375,000		29,167	5633	10,683

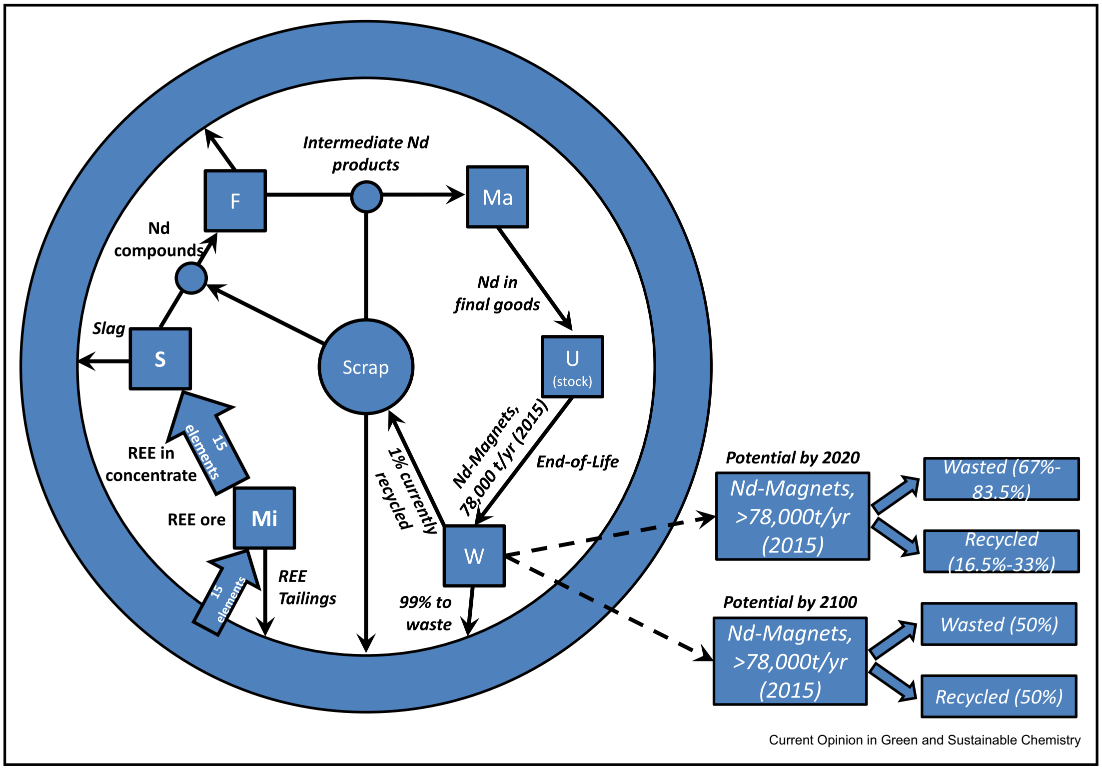
- Gas phase extraction methods where the REE are transferred to a volatile chloride phase and are separated based on differences in volatility.

The manufacture of NdFeB magnets also utilises a number of grinding, cutting and polishing operations that can lead to nearly a third of input REE being lost as manufacturing scrap [12]. Some present end-of-life uses mean that it is possible in some cases to re-use magnets in their current form. However, this is typically only possible for larger magnets used in electric vehicles or wind turbines that appear less commonly in waste streams as a result of their longer usage lifetimes [17]. This leaves waste electrical and electronic equipment (WEEE; notably computer hard disk drives and mobile phones) as a more valuable source of end-of-life REE-containing magnets. The pre-processing of WEEE involves the automatic sorting of NdFeB magnets, potentially resulting in crushed magnetic components being attracted to ferrous metal scrap. Although this behaviour can effectively separate the REE into a single output stream, the resulting mass concentrations are typically low and in most cases this approach results in the contamination of this stream [18], potentially meaning that the recovery of the REE from this recycling stream is not possible.

Fluorescent lamps

Energy-efficient and longer lasting compact fluorescent lamps have increased in use over the past two decades, leading to a coincident increase in demand for the REE in the form of REE (predominantly Y, Eu and Tb) phosphors (e.g., [19]). Typical fluorescent lamp waste contains >20% REE by weight (e.g., Refs. [19,20]) in the form of white, red, green and blue phosphors, each of which contain differing amounts of the REE (e.g., [12]). Recycling approaches include a) direct re-use, although this is only applicable in a very limited number of cases, b) the individual separation of phosphor components, a relatively simple approach but one that does not easily yield pure phosphor fractions, or c), chemical extraction of the REE, an approach that could yield individual REE for reuse in a variety of end-products but one that is much more difficult and energy and chemically intense than the other approaches (e.g., [16]) The majority of research to date has focused on the extraction of Y and Eu from red

Figure 1



Circular flow diagram of the Nd element cycle (adapted from Ref. [37] with information from Refs. [11,38–40]), showing the cycling of Nd from mining (Mi) thorough refining and separation of Nd from the other REE (S), the fabrication of Nd-bearing products (e.g., magnets; F), the incorporation of these Nd-bearing products into end-products during manufacturing (Ma), the use of these end-products (U), and the final disposal of these end-products to waste (W) at the end of their useful lives, with the resulting Nd either deporting to waste or being recycled. Two recycling scenarios are shown, the status quo, where very little Nd is recycled as discussed in the text, and future Nd recycling by 2100 where a significant proportion of Nd can be recycled. Text in italics relates to possible future sources of Nd and the other REE either as a result of recycling or by the increased recovery of these critical elements from waste products generated within this cycle.

phosphors as these elements are relatively easy to recover and are high value (e.g., [19]). However, the La-, Ce-, and Tb-bearing green phosphors are also an attractive target for future recycling, although these phosphors are harder to dissolve, requiring high temperature acid dissolution of molten NaOH or Na_2CO_3 approaches [19,21,22]. As emphasised by S Van Loy et al. [19], future research should focus on developing effective and efficient methods for the recovery of the REE from these green phosphors.

Catalysts

The REE are extensively used in catalysts employed during hydrocarbon cracking and contain around 3.5% by weight of La with lesser amounts of Ce, Pr, and Nd [12]. There has been very little interest to date in recycling the REE within these catalysts, with research focussing on acid leaching [12]. This may be related to the fact that the REE in these catalysts are relatively low value (e.g., Ref. [5]), making it unclear whether the recycling of the REE in these catalysts is currently or indeed will become economic.

Ni-MH batteries

Rechargeable nickel metal hydride (Ni-MH) batteries contain around 10% REE that are present in order to impart hydrogen storage capabilities [12,18]. These batteries use misch metals (alloys of the light rare earths generated by fused chloride electrolysis) or metallic stage LREE (predominantly La, Ce, Pr, and Nd) alloys. Until recently, the dominant recycling of these batteries was in stainless steel production as a cheap source of Ni, with the REE deporting to smelter slags and being lost [12,23]. However, the REE within these batteries could potentially be recycled using hydrometallurgical or pyrometallurgical recovery routes although both approaches are still in their infancy (e.g., [12]). In terms of the potential amounts of the REE that could be recovered from these (and other) batteries, Ueberschaar et al. (2017) [18] examined the apportionment of different elements during WEEE pre-processing. They determined that approximately one third of the REE in a typical WEEE waste stream could be manually sorted via the removal of batteries. Although this does not account for the majority of the REE contained in WEEE, the authors consider this to be a more attractive source for REE recovery as a result of a higher output mass fraction. However, P Sommer et al. [24] indicated that the low collection rates of batteries (i.e., in shavers, tooth brushes and cordless telephones) is a major inhibiting factor to the future recovery of these potential REE resources, again indicating that some of the inhibitors to REE recycling are logistical or infrastructural (e.g., Table 2).

Future developments in the recycling of the REE

The future potential for the recycling of the REE is prevalent on the type of material being recycled, and a significant amount of laboratory-based research has been undertaken to develop potentially scalable approaches that would enable the broader recycling of the REE. However, recycling of the REE currently requires extensive dismantling and the development of efficient collection infrastructure. As described above, another major issue blocking future developments in the recycling of the REE is the lack of cost effective methods to purify the mixtures generated during the recycling of consumer devices such as WEEE. Some recent advances in research in this area provide insights into approaches that can tackle this issue. For example, Bogart et al. [25,26] developed an organic compound that could bind to REE cations, enabling the formation of different compounds for the light REE (LREE) and the heavy REE (HREE), with the LREE forming dimeric aggregations whereas the HREE compounds not aggregating. This generated solubility differences that were large enough to allow the effective separation of the LREE and HREE by filtration [25,26]. The optimization of this separation allowed the formation of specific REE pairs that are useful in manufacturing, for example Nd-Dy and Eu-Y that are used in permanent REE magnets and fluorescent bulbs. This new approach to REE recycling, if scalable, may provide a significant step towards increasing the recyclability of a wide variety of REE-bearing end products.

It is important to also acknowledge that recycling is one among many possible responses to perceived REE supply risks. On the supply side, research and exploration should focus on the discovery of new mineral deposits, adapting existing mines to process the REE (e.g., monazite from heavy mineral sands mines; [27]), developing process technology for preferentially extracting the heavy REE (e.g., the Dubbo-Toongi project in Australia), as well as recycling. The potential for REE extraction from secondary sources such as low-grade REE industrial residues (e.g., phosphogypsum, slags, bauxite residue (red mud), mine tailings, metallurgical slags, coal ash, incinerator ash and waste water streams) is also significant [12]. Studies like S Peelman et al. [28] have highlighted the potential for the extraction of the REE from mine tailings, in this case from the Kiruna iron ore mine in Sweden. Recovering the REE from acid mine drainage could also have economic, environmental, and strategic benefits (e.g., [29,30]). Equally, research into other methods of recycling should be promoted; for example, bioleaching has a much lower environmental footprint compared to other REE recycling methods (e.g., Refs. [2,31–33]).

On the demand side, research and development should focus on finding substitutes for the REE in different technologies or the finding of alternatives for these technologies altogether (if possible). While these latter approaches may seem less likely given generally low substitutability ratings [34], the substitution of Nd in NdFeB magnets was in fact a leading response to the so-called 2010 "rare earth crisis", fostering a return to pre-crisis prices for this metal [35]. Indeed, tech companies like General Electric have been known to perform criticality assessments to reduce their dependence on the REE and other critical metals [36]. We can expect that the future recycling rates of the REE will thus be significantly influenced by the external efforts of both mining companies and manufacturers.

Conclusions

The REE are among the most critical of the critical elements yet current efforts to recycle these valuable commodities are seemingly relatively ineffective. There is significant potential to increase the amount of the REE recycled from major end-uses, such as permanent magnets, fluorescent lamps, batteries, and catalysts; however, a significant amount of research is needed in all of these areas to increase the amount of these elements. Increased amounts of REE recycling has the potential to play a key role in addressing a number of criticality issues with these elements, including meeting increased demand, increasing their security of supply, and overcoming the balance problem between higher and lower demand REE and the concentrations of the REE available from primary mine-derived sources.

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