

Review

Sourcing, Refining and Recycling of Rare-Earth Magnets

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Abstract: Permanent magnets today are used in a wide range of transportation, industrial, residential/commercial, consumer electronics, defense, domestic, data storage, wind energy, and medical markets and applications. There are five classes of commercial permanent magnet materials; however, magnets based on Nd-Fe-B account for over 60% of the global magnet production by value. They typically contain around 31 wt.% of rare earth elements (REEs), principally, Nd and Pr, plus Dy for higher-temperature performance. Nd-Fe-B magnets are forecast to grow throughout this decade, largely driven by the growth in electric vehicles of all types. However, several studies forecast a shortfall of the primary REEs from mined resources. In this paper, the sourcing, processing, and recycling of REEs are discussed. Additionally, presented are the advantages and disadvantages of the major recovery and recycling technologies for REEs.

Keywords: rare earth elements; magnets; Nd-Fe-B; electric vehicles; motors; recycling

1. Rare-Earth Elements (REE): Sources, Classification, Concentration, and Refinement

Rare-earth elements (REEs) are a group of 15 elements in the periodic table, ranging from lanthanum to lutetium, also known as lanthanides, plus scandium and yttrium. Depending on their atomic numbers, REEs can be separated into light rare-earth elements (LREEs) and heavy rare-earth elements (HREEs). LREEs include lanthanum, cerium, praseodymium, neodymium, samarium, and europium, and HREEs are defined by the suite of lanthanides that include gadolinium, terbium, dysprosium, holmium, erbium, thulium, ytterbium, and lutetium, with those elements most used in high-performance permanent magnet applications highlighted with horseshoe magnet symbols in Figure 1 [1–5].



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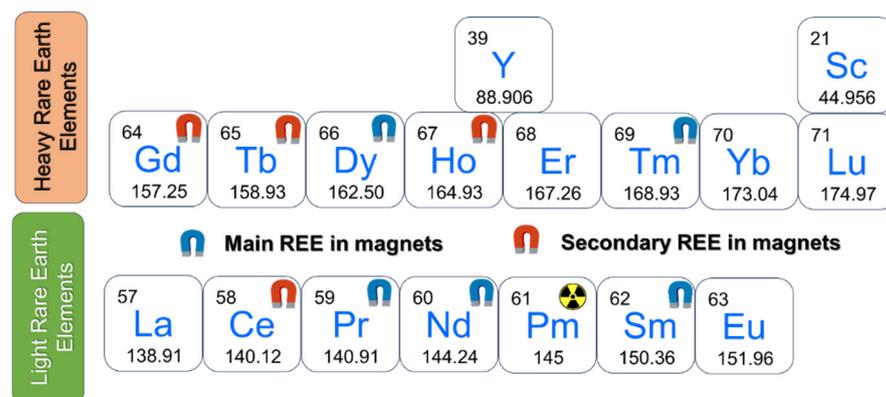


Figure 1. Classification of REEs based on their application in magnets.

The REE resources can be broadly classified into primary and secondary sources. Primary REE sources are predominantly mineral ores produced by magmatic, hydrothermal, or metamorphic processes [6]. Commercial extraction of REEs is dominated by a few

mineralogies, including bastnaesite, ion-adsorption clays, monazite, and xenotime, which account for over 95% of economic production. The LREEs are predominantly recovered from mineral concentrates of monazite and bastnaesite sourced from operations in China, the USA, Australia, India, and Madagascar, and to a lesser extent from the mineral loparite extracted in Russia. Most HREEs are produced from ion-adsorption clays and xenotime mineralization in Southern China, Myanmar, and Australia. Though xenotime is less abundant than other minerals, it is a primary source of HREEs, particularly dysprosium. Apatite has also been mined in its non-weathered state for REEs [1]. Table 1 shows a list of naturally occurring REE-containing ores. Bastnaesite, iimoriite, monazite, parisite, and xenotime are the ones that contain the maximum fraction of REEs [7].

Table 1. REEs containing natural ores [7].

Mineral	Formula	REO (wt. %)
Allanite	$(Y, Ln, Ca)_2(Al, Fe^{3+})_3(SiO_4)_3(OH)$	39
Apatite	$(Ca, Ln)_5(PO_4)_3(F, Cl, OH)$	19
Bastnaesite	$(Ln, Y)(CO_3)F$	75
Eudialyte	$Na_4(Ca, Ln)_2(Fe^{2+}, Mn^{2+}, Y)ZrSi_8O_{22}(OH, Cl)_2$	9
Fergusonite	$(Ln, Y)NbO_4$	53
Iimoriite	$Y_2(SiO_4)(CO_3)$	68
Kainosite	$Ca_2(Y, Ln)_2Si_4O_{12}(CO_3) \cdot H_2O$	38
Loparite	$(Ln, Na, Ca)(Ti, Nb)O_3$	30
Monazite	$(Ln, Th)PO_4$	65
Mosandrite	$(Na, Ca)_3Ca_3Ln(Ti, Nb, Zr)(Si_2O_7)_2(O, OH, F)_4$	33
Parisite	$Ca(Ln)_2(CO_3)_3F_2$	61
Rinkite	$(Ca, Ln)_4Na(Na, Ca)_2Ti(Si_2O_7)_2(O, F)_2$	20
Steenstrupine	$Na_{14}Ln_6Mn_2Fe_2(Zr, Th)(Si_6O_{18})_2(PO_4)_7 \cdot 3H_2O$	31
Synchysite	$Ca(Ln)(CO_3)_2F$	51
Xenotime	YPO_4	61
Zircon	$(Zr, Ln)SiO_4$	4

Secondary REE sources, on the other hand, originate from industry byproducts that otherwise end up in landfills owing to their dilute concentrations [8,9]. Furthermore, secondary sources of REEs also include electronic wastes and several other useful commercial commodities. Thus, while the idea of primary REE sources is clear, the definition of secondary sources must be clarified for consistent referencing. In the present report, we thus broadly classify the secondary REE sources into two major categories, namely: (a) unprocessed and (b) processed sources. The unprocessed sources are REE-containing materials without any prior commercial use. They include mine tailings, coal ash, phosphogypsum, red mud, and marine sediments [10]. Conversely, the processed sources relate to REEs that have already been used for certain applications. These include REEs in catalysts, permanent magnets, polishing materials, fluorescent and LED lamps, metal hydride batteries, and electrical appliances and other applications [11].

REE sources are also categorized based on conventional and unconventional sources. While ores containing REEs have been strictly categorized as conventional sources, mine tailings and byproducts from coal ash, apatite, and phosphates have been labeled unconventional REE sources [12]. E-waste generation, continually rising over the years, is also seen as an unconventional REE source. However, with the fast depletion of conventional REE sources, unconventional sources may become the conventional and primary sources.

Furthermore, the primary REE sources are sparsely dispersed worldwide. A recent US Geological Survey report suggests that four countries possess more than 85% of the world's natural REE resources [13] (Figure 2, Table 2). Figure 3 shows a global map of countries with REE reserves. Countries such as the United States are already mining their REEs at an unprecedented rate, which might bring forth the REE crisis in the coming decades [13] (Figure 2, Table 2). This has thus brought forth the idea of urban mining, which looks at recycling e-wastes to recover rare-earth resources as an alternative approach [14].

“Mining” REE from e-wastes can be more environmentally friendly than mining from virgin resources [15]. In the present report, we suggest demarcating REE reserves into two categories: primary and non-primary. The non-primary resources would include all the secondary and unconventional REE resources.

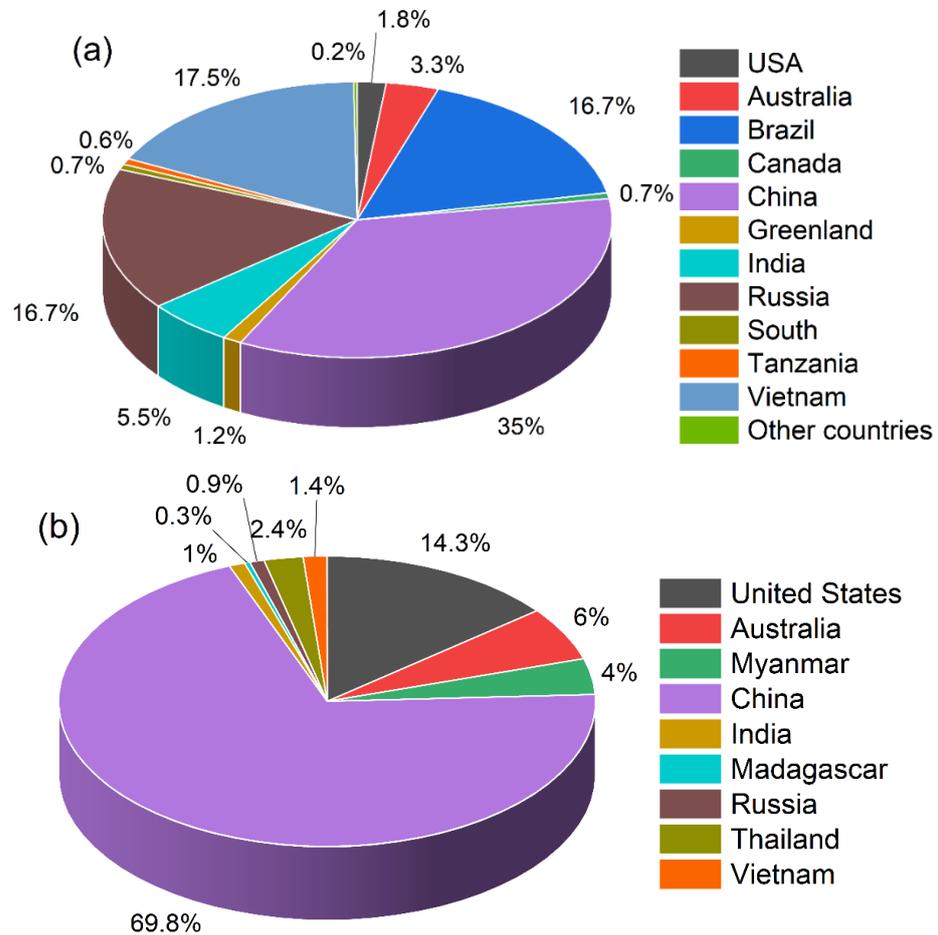


Figure 2. Share of (a) REE reserves and (b) mined REE worldwide. Data for the plot were extracted from [13].

Table 2. REE reserves for different countries as of 2022 [13].

Country	Mine Production (Tons)	Total Reserves (Tons)	% of Reserves Mined	Share of World Reserves
United States	43,000	2,300,000	1.87	1.83
Australia	18,000	4,200,000	0.43	3.34
Brazil	80	21,000,000	0.0004	16.71
Myanmar	12,000	-	-	-
Canada	-	830,000	-	0.66
China	210,000	44,000,000	0.48	35.01
Greenland	-	1,500,000	-	1.19
India	2900	6,900,000	0.04	5.49
Madagascar	960	-	-	-
Russia	2600	21,000,000	0.01	16.71
South Africa	-	790,000	-	0.63
Tanzania	-	890,000	-	0.71
Thailand	7100	-	-	-
Vietnam	4300	22,000,000	0.02	17.50
Other countries	80	280,000	0.03	0.22

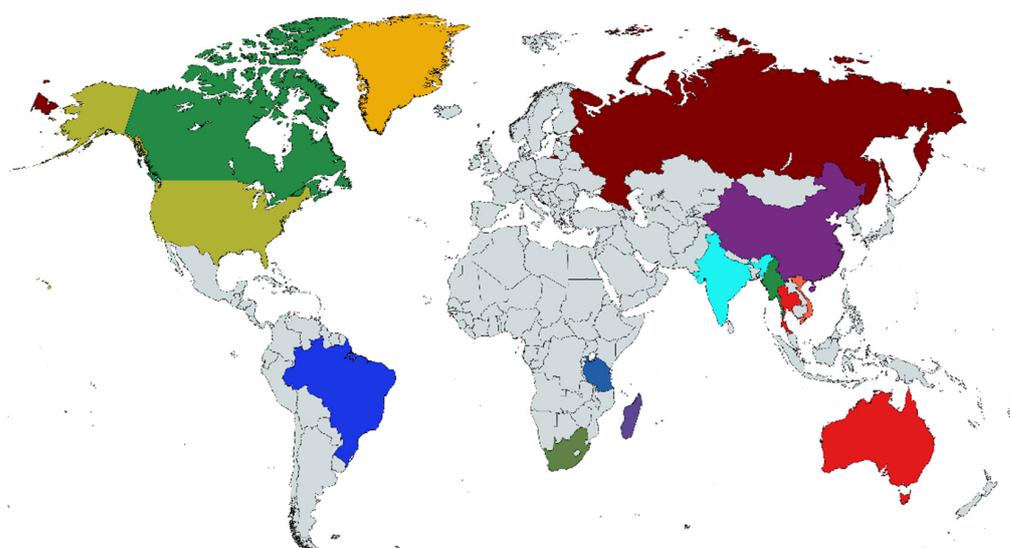


Figure 3. World map showing countries with REE reserves as of 2023. Non-grayscale colors have no other significance than to highlight the countries indicated.

Several projects are currently assessing REE from various unconventional sources that will propel countries into more self-reliance and help alleviate their need to rely on imports of such materials. Table 3 briefly lists key activities from coal byproducts and acid mine drainage to extract REEs.

Table 3. Key activities investigating the extraction of REE from coal-based products and acid mine drainage.

S. No.	Institution	Key Goals and Findings
1	University of Illinois, USA	A database of carbon ore, rare earth, and critical minerals (CORE-CM) has been developed by collecting datasets from several thousand samples along the Illinois Basin. Development of technology to aid in separating RE materials operating at or near the mine-face. [16]
2	University of Kansas Center for Research, USA	The commissioning of a Geotek core scanner will assess CMs in the Cherokee forest City Basin. This includes regions of Kansas, Iowa, Missouri, Nebraska, and Oklahoma, and Osage Nation. [17]
3	Collaborative Composite Solutions Corporation, USA	Revitalize the coal production for CM assessment in the Southern Appalachian Basin. Parameters needed to determine REE security costs for the US in the coming years have been investigated. [18]
4	University of Texas, Austin, USA	Investigation of REE from coal mines and power plants in the US Gulf Coast Basin. Investigations found significant REE in Gibbons Creek mine (TREE: 1000–8000 ppm) and San Miguel mine (TREE: 300–900 ppm). [19]
5	NETL, USA	Investigation of CORE-CM in Usibelli Coal Mine and Graphite Creek in Alaska. Preliminary studies have indicated an encouraging amount of REE minerals in coal samples from the mines. [20]
6	LANL, USA	Quantitative investigation of REEs in coal samples using laser-induced breakdown spectroscopy (LIBS). Construction of LIBS in a backpack to quantitatively analyze the presence of REE in coal. [21]

Table 3. Cont.

S. No.	Institution	Key Goals and Findings	
7	West Virginia University, USA	Development of pilot-scale plant for treating acid mine drainage to produce RE and CMs. Extracted and separated samples demonstrate the presence of >67% HREE+CM. 807 tons of TREE production per year waste product is non-hazardous and can be disposed of on-site.	[22]
8	Florida Polytechnic University, USA	Extract REE from phosphoric acid sludge. Demonstrated extraction of 90% REE and 100% heavy REEs. Planned production capacity of 900 to 1100 tons of REM per year.	[23]
9	Institute of Environmental Assessment and Water Research, Spain	7.9 and 3.5 mg/L REE and Y concentrations obtained from Monte Romero and Almagrera acid mine drainage. REE includes predominantly La and Ce.	[24]
10	Institute of Earth Sciences, Germany	1.8 and 2.5 mg/L concentrations of REE detected in Giessenbach Creek. Higher concentrations of middle and heavy REEs were detected.	[25]
11	Universidade de Aveiro, Portugal	110, 120, and 124 mg/kg of REE located in Lousal mine area in the Iberian Pyrite Belt, Portugal. 14–20 mg/kg of HREE detected.	[26]
12	IIT Kharagpur, India	0.71 mg/L of REE detected in mine drainage from Jaintia Hills coalfields, India.	[27]
13	University of Huelva, Spain	Origin of REE traced in acid mine drainage from the Iberian Pyrite Belt (SW Spain). 20–30 mg/kg of REE detected in felsic volcanic from Poderosa mine. 20–30 mg/kg of REE detected in Shales from the Perrunal mine.	[28]

Concentrates of REE minerals are exclusively processed into refined products, either as mixed/semi-separated compounds or individual rare-earth compounds. Most refining occurs within China, Malaysia, Russia, and India, with small amounts separated in Vietnam, Norway, and Australia. Refined REEs have a range of end uses, but increasingly, high-powered permanent magnet applications dominate the sales value of REEs. Today, the REEs used in permanent magnets (Nd, Pr, Dy, and Tb) account for over 90% of the value of all REEs processed [6].

Over 50% of REE demand has historically come from China, other Asian countries, Australia, and North America. Under this context, global demand for REEs increased from below 157 kt of rare-earth oxide (REO) in 2017 to 256 kt REO in 2020, expecting to reach 305 kt REO by 2025 [29]. Due to the 2019 COVID-19 pandemic and its effects, the REE market contracted by 1% in 2020; however, its overall increase in demand has seen REE consumption reach record highs, enhanced mainly by rapid growth in permanent magnet applications [30,31]. In 2022 and early 2023, China still accounted for 70% of REE production and 90% of processing [32]. In general, REEs' demand will be increasingly driven by their use in permanent magnets for the electrification of transport and the transition to renewable energy generation. This is evident from the more than 40% increase in refined REEs for permanent magnets between 2020 and early 2023 [32]. Simultaneously, the transitioning away from fossil fuels in transport and energy generation will reduce the REE demand from catalyst applications in both petroleum processing and emissions control systems for passenger and commercial vehicles [33].

Permanent magnet applications play a critical role in supporting the energy transition. Transport, energy-efficient equipment, and energy generation applications will cause significant growth in the demand for key magnet elements, with Nd and Pr being the most impacted, as they are the main rare-earth constituents of Nd-Fe-B alloys by volume. Short-term demand growth for Tb and Dy will accelerate in the period to 2030 before slowing, as the development of new magnet designs and production methods reduce the required HREE intensity in high-performance magnets. Replacement of some Nd-Pr by La

and Ce tends to reduce the performance of the magnets, which may be unsuitable for the highest growth markets, such as automotive drivetrains. Using La and Ce in such mid- to low-quality magnets can increase their demand in the future.

Once mined, RE minerals are processed into concentrates containing multiple REEs, which need to be separated from each other (Figure 4), typically by solvent extraction. REEs are chemically very similar, so separation often requires a series of extractions using multiple solvents to separate desired individual or compound REEs. This step in the process involves large amounts of acid, water, and radioactive byproducts, so obtaining adequate solvents and treatment of waste are significant cost drivers. REOs must be further refined or reduced to metal before they can be used for magnet production (Figure 4). Electrowinning is the most common process for converting REOs into their metallic state, while ensuring low-impurity contents, particularly oxygen, nitrogen, and carbon [33,34].

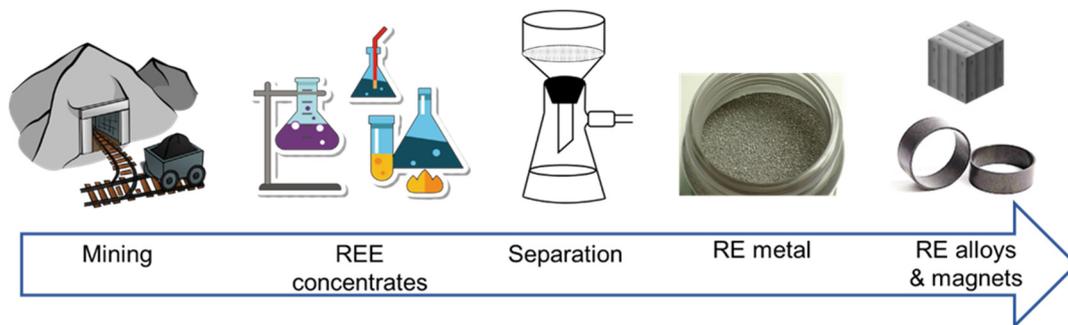


Figure 4. Steps in the processing and refinement of REEs.

2. Rare-Earth Permanent Magnets and Applications

The special technological importance of permanent magnets derives from their ability to produce a magnetic field, making them suitable for various transportation, industrial, residential/commercial, consumer electronics, defense, information technology, power generation, and medical applications. Unlike electromagnets that require a continuous electrical current to be supplied to generate a magnetic field and function as a source of magnetic flux [31,35,36], permanent magnets provide a magnetic flux with no external energy input.

Not all permanent magnets utilize REEs. There are four general classifications of permanent magnets, each with a range of performance metrics and typical end-uses, and these include ferrite, alnico, samarium-cobalt (Sm-Co), and neodymium-iron-boron (Nd-Fe-B). Nd-Fe-B-based magnets are the backbone of expansion in alternative energies, although their performance at high operating temperatures is limited. This limitation is typically overcome by adding Dy or Tb, which increases the coercivity and performance at higher temperatures. Several extensive reviews are available that cover the history of REE permanent magnet development [37–43]. Nd-Fe-B magnets typically contain around 32 wt.% of REEs, principally, Nd and Pr, plus Dy and Tb for higher-temperature performance. Figure 5 shows the estimated global production of permanent magnets by material type [31].

Together, the ferrite- and Nd-Fe-B-based magnets account for over 90% of the global production of permanent magnets. Despite the significant difference in magnetic performance between the two materials, their respective price-to-magnetic performance ratio is extremely favorable compared to all other material options. The high energy density of Nd-Fe-B makes them the magnet of choice for many energy-efficient and renewable energy applications, e.g., EV drivetrain motors, wind power generators, and HVAC (heating, ventilation, and air conditioning) units. Consequently, the market for these magnets is forecast to grow from 200,000 tons per year in 2022 to 450,000 tons in 2030 [31]. The details, by major markets, are shown below in Figure 6.

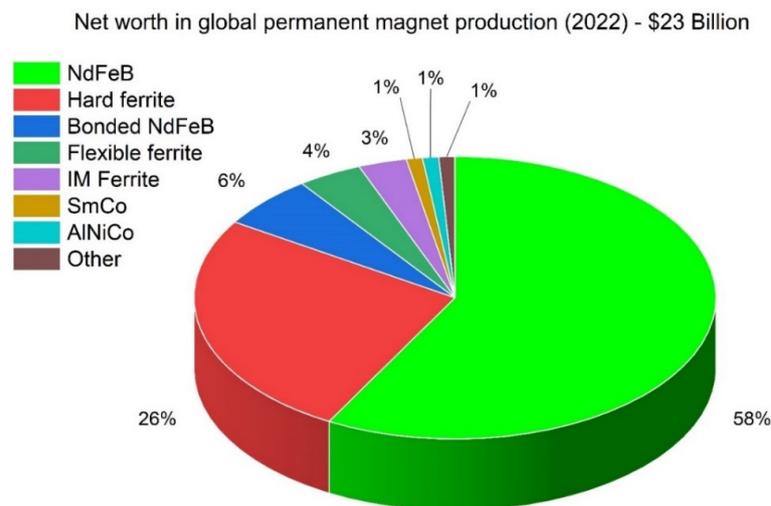


Figure 5. Global permanent magnet production by value in 2022 [31].

As can be seen, the growth in demand is largely driven by renewable energy applications. Unfortunately, the major REEs used, namely Nd and Pr, and for higher-temperature applications, Dy and Tb, are forecast to be in short supply, considering these aggressive demand forecasts. For example, the Adamas Intelligence forecast indicates that by 2040, the undersupplies of Nd-Pr, Dy, and Tb oxides will reach 90 kt, 1.8 kt, and 0.45 kt, respectively [44]. Nd-Fe-B magnets are forecast to grow at a CAGR of over 10% throughout this decade, largely driven by the growth in electric vehicles of all types. Several market studies are predicting that combined production of electric vehicles and hybrid electric vehicles (EVs/HEVs) will be in the range of 50 to 100 million units produced annually by 2030. Current EV/HEV motors contain, on average, 1.8 kg of Nd-Fe-B-based magnet material per vehicle [45]. This equates to over 90,000 mt of Nd-Fe-B magnet material at the low end of the forecast consumption for this one application in 2030. The preferred motor type used in EV traction drives is the interior permanent magnet (IPM) design. In this design, the magnets are installed into slots in the laminated rotor [45].

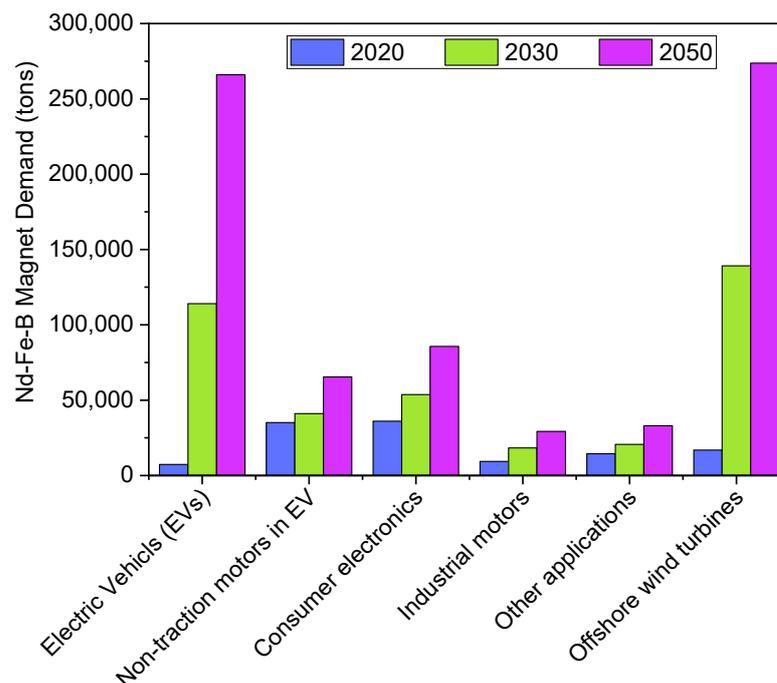


Figure 6. Major growth markets for Nd-Fe-B magnets through 2030. Adapted from [45].

It is generally accepted that the magnetic parameter, maximum-energy product $(BH)_{max}$, is the best all-round single indicator of permanent magnet performance. From the first principles, it can be shown that for a magnetic circuit containing an airgap, the energy stored in the field in the air gap is directly proportional to the product of flux density, B , and the corresponding field strength, H , at any point on the second-quadrant normal demagnetization curve, as shown in Figure 7 below [46]. The maximum value of this product, i.e., $(BH)_{max}$, or the maximum-energy product, can be directly related to the maximum energy that can be generated with a permanent magnet [47].

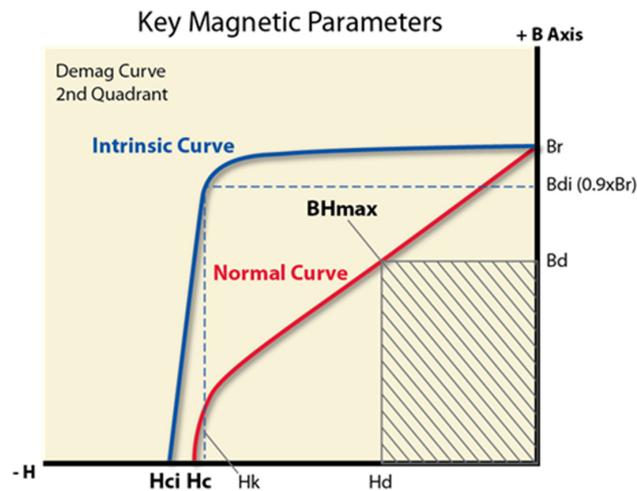


Figure 7. Second–quadrant demagnetization curves [46].

The following chart (Figure 8) shows the historical development and commercialization of permanent magnets based on their maximum-energy product, $(BH)_{max}$. This chart clearly demonstrates the improvement in magnetic performance, beginning with magnet steels in the early 1900s, to alnico's, and finally, hard ferrites and rare-earth magnets. It is now over 40 years since the US announcement of Nd-Fe-B magnets at the 29th MMM conference held in Pittsburgh, PA, in November 1983 [22]. It is interesting to note that since the discovery and introduction of Nd-Fe-B magnets, no major new sintered permanent magnet material has been introduced.

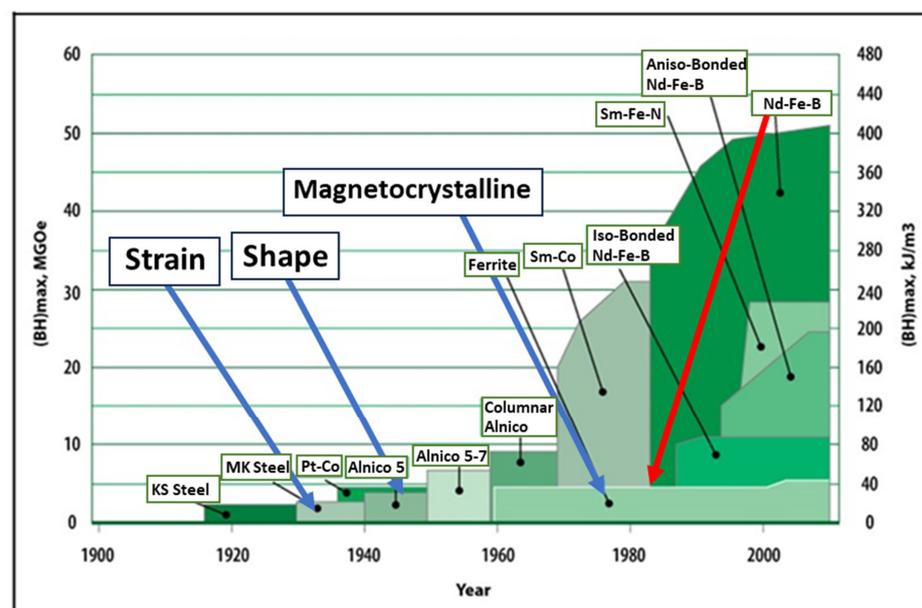


Figure 8. Historical development of permanent magnets [48].

3. Sintered Nd-Fe-B-Based Magnet Processing

Figure 9 outlines the basic steps for the powder metallurgical processing of sintered Nd-Fe-B-based magnets [49,50]. The metallization step involves the production of the REE metals through the reduction (mostly electrolytically) of RE_2O_3 (RE = Nd, Pr, and Dy, or their mixtures). Electrolytic reduction is accomplished by passing electric current through electrodes in a molten mixture of the oxide and a fluxing agent. Keeping the oxygen, nitrogen, sulfur, and carbon levels low is an important consideration for the subsequent permanent magnet production because removing any impurity later in the process is impractical.

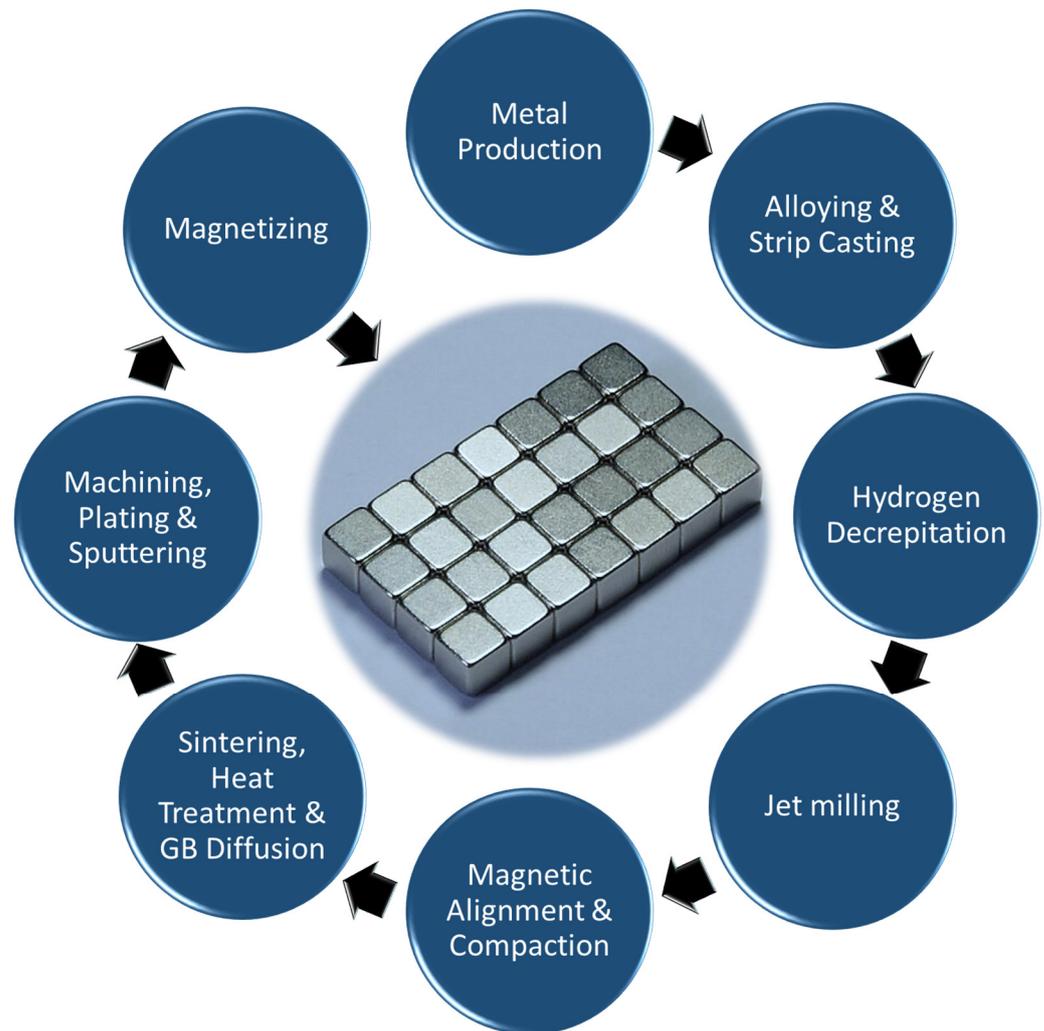


Figure 9. Powder metallurgical processing of Nd-Fe-B-based magnets [49,50]. Grain boundary is abbreviated as GB.

The next step is the preparation of the master alloy using Nd, Pr, and Dy metals (or their alloys), along with iron, cobalt, ferro-boron, and in many cases, reclaimed and recycled alloy material. Other additive elements (Al, Cu, Ga, etc.) are included to enhance grain boundary refinement. The master alloy is subsequently melted and rapidly cooled into flakes, preferably via the strip-casting process (Figure 10). This rapid cooling results in the right phase and microstructure for developing the optimum magnetic properties in the finished magnet. This is critically important because slow cooling favors the formation of α Fe, which makes crushing and grinding the ingot into a powder much more difficult and deteriorates the magnetic performance of a permanent magnet [51]. How to eliminate the formation of α Fe is part of the lessons learnt in the early stages of the development of

sintered Nd-Fe-B magnets. At the time, conventional metal molds were used to cast the alloys, resulting in relatively slow cooling rates and significant α Fe formation. The solution was the development of the strip-casting process for Nd-Fe-B alloys.



Figure 10. Image of typical alloying and strip-casting equipment.

The next step is the crushing of the strip-cast alloy to a coarse powder. Today, this typically uses the hydrogen decrepitation process, which is shown diagrammatically in Figure 11 [49]. In this step, the alloy is exposed to hydrogen gas pressure, during which it absorbs hydrogen in the grain boundaries, resulting in decrepitation into coarse powder. The coarse alloy powder is afterwards subjected to jet milling, during which it is comminuted to a fine powder with an average particle size of 5 μm (Figure 12). The objective of jet milling is to produce a powder consisting of single-crystal particles with a narrow particle size distribution. A typical jet milling system used for Nd-Fe-B magnets is schematically represented in Figure 13 [52].

Most commercially available sintered magnets are anisotropic, i.e., the magnetic easy axes of the powder particles (ideally single crystalline) are aligned in one direction. This helps to achieve a high remanent magnetization (hence, high $(BH)_{max}$) and a more coherent rotation of the magnetic moments during demagnetization. For sintered magnets, this is accomplished by applying a magnetic field and compacting the powders in the magnetic field, such that the easy axes of magnetization are parallel. Figure 14 depicts a typical pulsed magnetizer equipment. The green body thus prepared is then ready for densification by sintering.

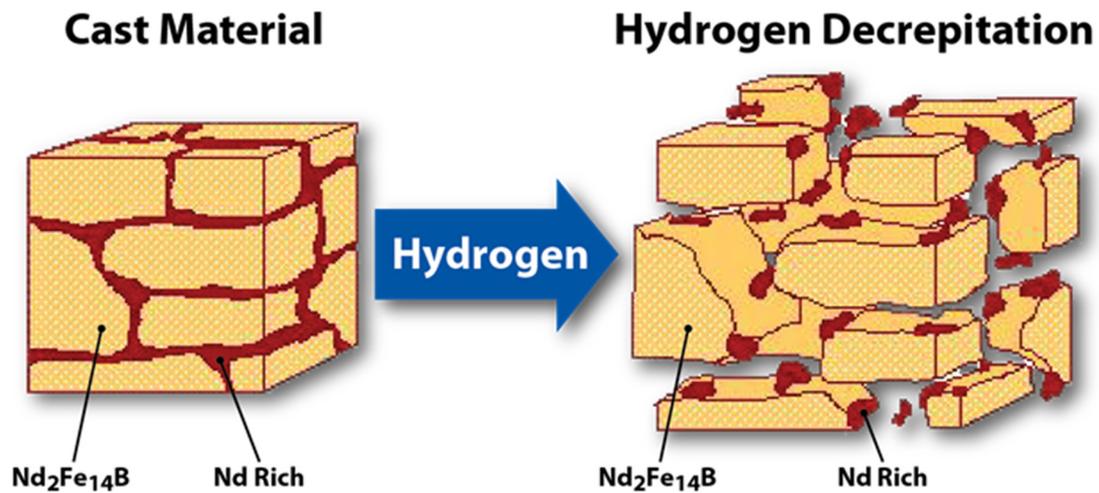


Figure 11. Schematic of hydrogen decrepitation of strip-cast Nd-Fe-B alloy [52].



Figure 12. Image of typical jet-milling equipment.

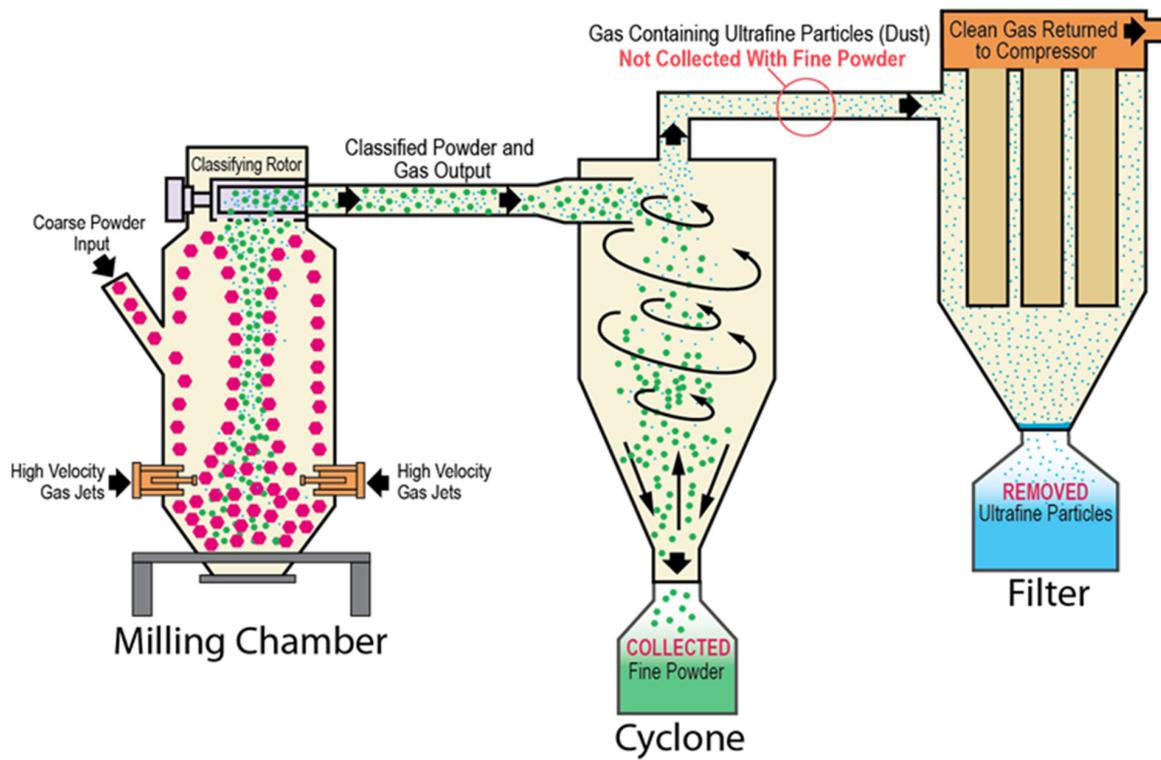


Figure 13. Schematic of a jet milling process [52].



Figure 14. Image of typical pulsed magnetizer equipment.

The sintering of Nd-Fe-B permanent magnets is typically undertaken in a vacuum furnace under a partial pressure of an inert gas such as argon. The vacuum furnace may need to be purged with an inert gas to minimize any oxygen and moisture contents. It is important to maintain a constant and well-controlled sintering temperature to ensure the magnet has no open porosity, as well as to prevent oxidation and aging during use. This requires the sintered density of the Nd-Fe-B magnet to be greater than 98% of the theoretical density.

A final thermal treatment is applied to the sintered Nd-Fe-B magnets to develop the microstructure for good hard magnetic properties. The addition of additive elements such as Al, Cu, and Ga (or their alloys) during the Nd-Fe-B alloy preparation is critical for this stage. For this, the Nd-Fe-B magnet alloys are prepared to be RE-rich, i.e., $RE_{2+x}Fe_{14}B_{1+y}$, such that the excess RE combines with the additive elements to form low melting (typically eutectic) phases in the grain boundary. The final thermal treatment leverages the low melting temperature (500–700 °C, depending on the constituent REs) of the phases to optimize the grain boundary for an enhanced hard magnetic performance, without significant grain growth. Grain growth deteriorates the coercivity of Nd-Fe-B, especially if non-uniform grain growth occurs. The shrinkage that occurs as part of the densification during sintering results in some magnet size variation, and therefore, final machining operations are necessary to achieve the required dimensional size and tolerances. Most times, magnets of smaller dimensions are machined from a larger sintered block.

Despite this commercial success, Nd-Fe-B magnets suffer from significant problems, including a high thermal coefficient of coercivity, β , high thermal coefficient of remanent magnetization, α , and poor corrosion resistance. β and α represent the percentage change in coercivity and remanence, respectively, with temperature, and are critical performance parameters. For Nd-Fe-B magnets, β can be up to $-0.65\%/^{\circ}C$, compared to $-0.3\%/^{\circ}C$ in Sm-Co magnets and $-0.01\%/^{\circ}C$ in alnico magnets. α in Nd-Fe-B magnets is also higher ($-0.1\%/^{\circ}C$), compared to $-0.05\%/^{\circ}C$ in Sm-Co magnets and $-0.02\%/^{\circ}C$ in alnico magnets. The negative sign is used to indicate a decrease in the properties; hence, the lower the absolute value, the better. The higher absolute values of β and α limit the maximum operating temperature of the Nd-Fe-B in application. Combining improved processing, microstructure refinement, grain boundary modification, alloy chemistry, and hermetic coatings has virtually eliminated the corrosion resistance issue. However, it is interesting to note that today's solution for increasing the operating temperature of a Nd-Fe-B magnet was discovered in 1984; namely, the addition of HREE elements and, in particular, Dy and Tb (see Figure 15). As can be seen, a small addition of Dy significantly increases the intrinsic coercivity, H_cJ , compared to the standard magnet composition [53]. The increase helps to ensure that there is sufficient coercivity for elevated-temperature applications.

Figure 16 shows an example of the demagnetization behavior of sintered Nd-Fe-B grade N55M (no Dy) at different temperatures. Note that both remanence and coercivity decreased with the increasing temperature. This grade loses around 50% of the room temperature coercivity at 100 °C, a major concern in any device operating at elevated temperature. Therefore, it has become common practice in the Nd-Fe-B magnet industry to add Dy or Tb to increase the intrinsic coercivity. Figure 17 shows the typical Nd-Fe-B grade designations developed based on Dy content and the corresponding maximum recommended operating temperature. It shows that increasing the Dy content increases the maximum operating temperature from around 80 °C to 220 °C.

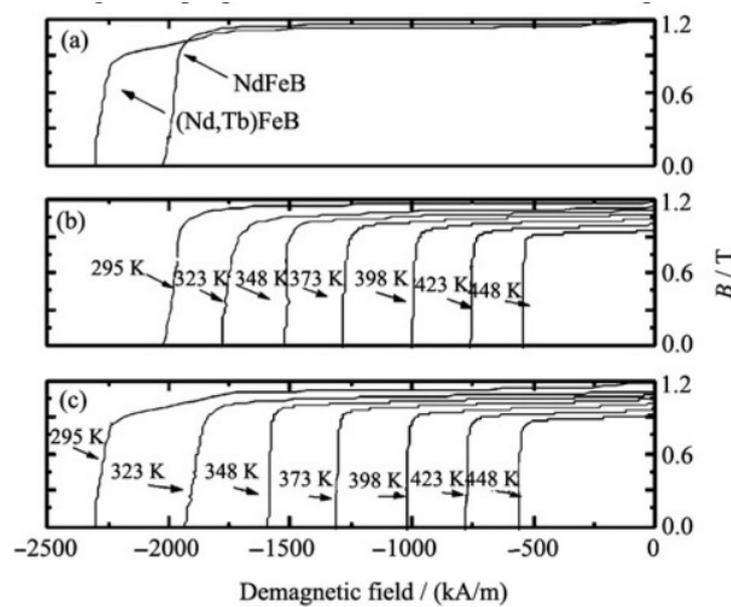


Figure 15. Effect of HREE (Tb) substitution on the coercivity of Nd-Fe-B magnets as a function of temperature. Demagnetization curves of Nd-Fe-B and (Nd,Tb)-Fe-B at room temperature (a); Temperature dependence of the demagnetization plots of Nd-Fe-B (b) and (Nd,Tb)-Fe-B (c). Reprinted (adapted) with permission from [53]. Copyright 2010 Elsevier.

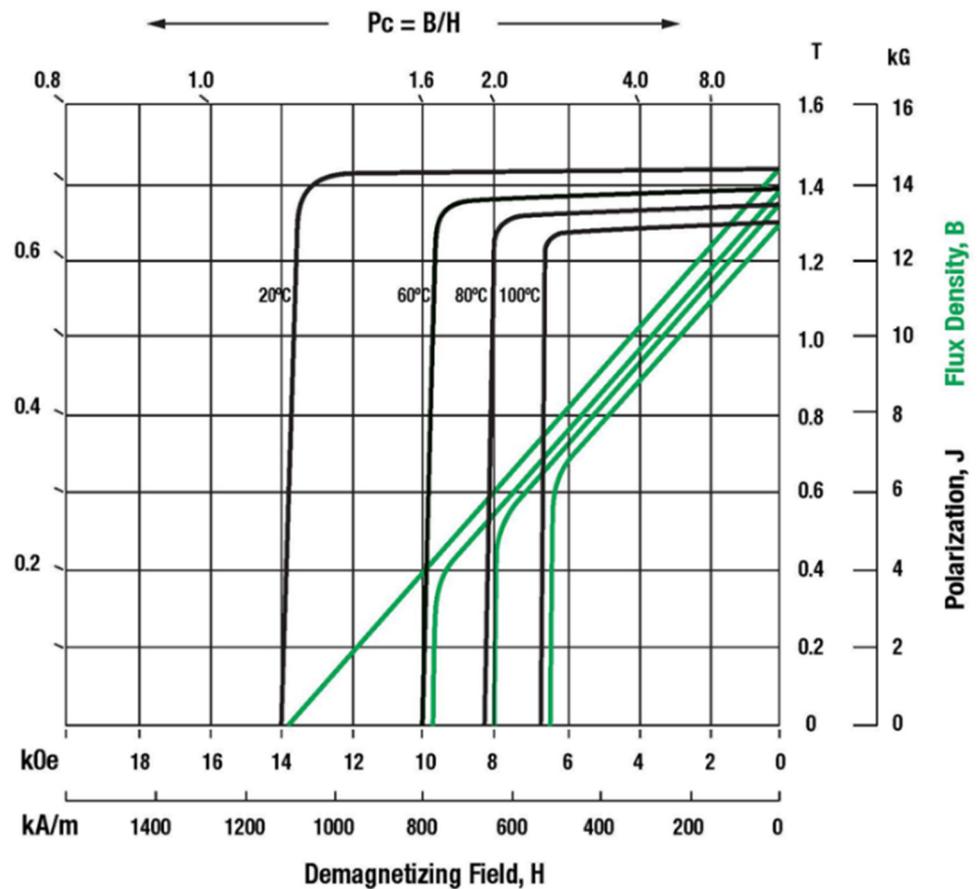


Figure 16. Demagnetization behavior of a N55-grade Nd-Fe-B magnet [54].

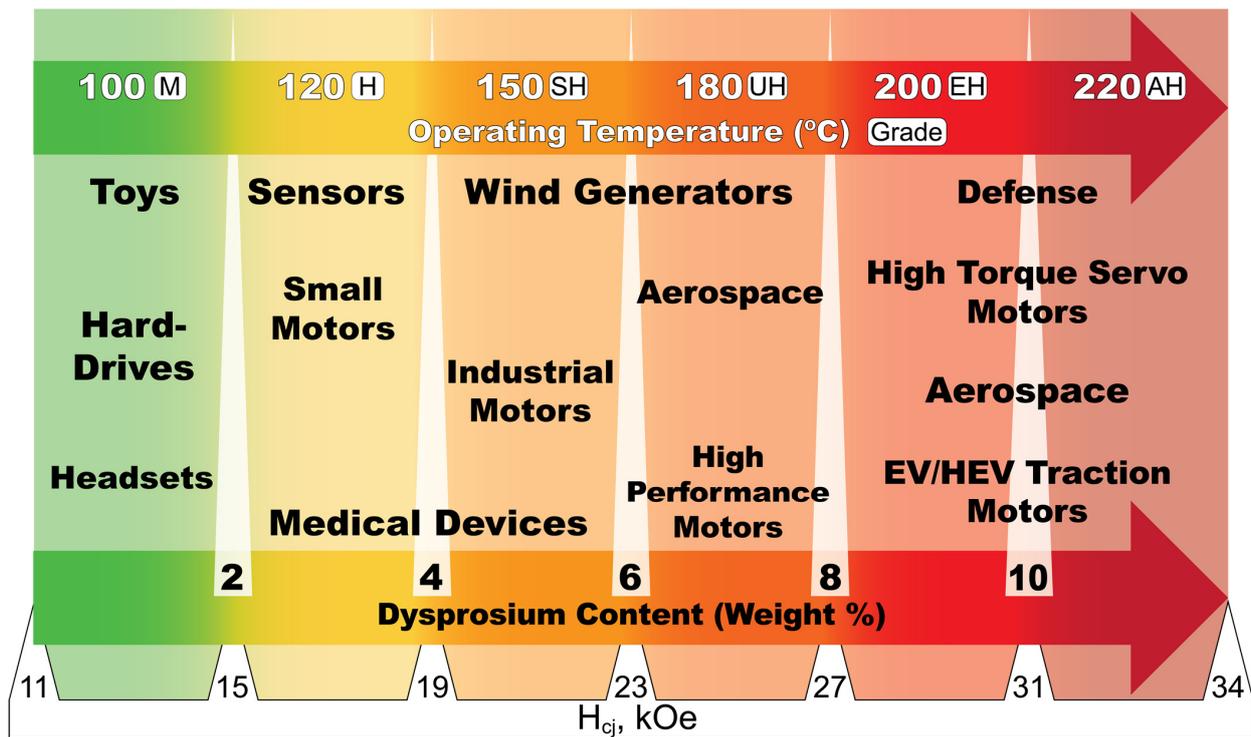


Figure 17. Nd-Fe-B grades and corresponding HREE content [55].

Following the severe disruptions to the REE supply chain between 2011 and 2012, extensive R&D efforts were undertaken to find processing methods for minimizing the Dy content for higher-temperature and high-coercivity Nd-Fe-B grades. A major advancement is to diffuse Dy from the surface of a sintered magnet, instead of alloying it in the entire magnet volume (Figure 18). The goal is to increase the anisotropy field, and hence coercivity, by enriching the regions near the grain boundary of the main $\text{Nd}_2\text{Fe}_{14}\text{B}$ phase with Dy. One of many variants is the TDK HAL (high anisotropy field layer) process [56]. In the HAL process, the Dy source is located on the surfaces of the sintered magnets and a low-temperature heat treatment under vacuum is applied, resulting in the solid-state diffusion of Dy around the grain boundary regions of the sintered magnet. This is shown diagrammatically in Figure 15. TDK claims the HAL process results in a 20% to 50% reduction in Dy content, with a 3% to 5% increase in Br. Today, it is common to find grain-boundary-diffused grades offered by all the major Nd-Fe-B manufacturers.

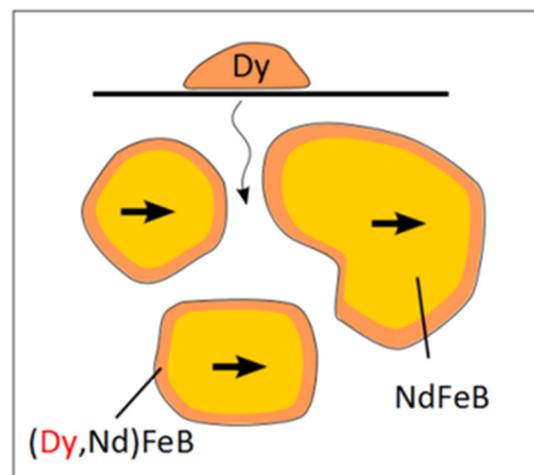


Figure 18. Schematic of the grain boundary Dy diffusion process [49,50].

4. Introduction to REE Recycling

Some amount of recycling has been part of manufacturing Nd-Fe-B since its commercialization in the mid-1980s. Going forward, the key will be the development of economically viable EOL magnet recovery and recycling processes. Today, this is a minor contribution to the total REE supply, but it needs to grow rapidly throughout this decade to become an important source of REEs.

Figure 19 shows the electric motor in a 2022 BMW X5 PHEV, which can be seen to be embedded in the drive train and surrounded by an aluminum alloy housing [57]. Economically removing the motor and extracting the magnets embedded in the rotor structure is not a trivial matter. There are some innovative technologies that can cost-effectively recover the REE compounds in magnets. Extraction of the magnets from EOL for reuse (as opposed to the recovery of the REE compounds) has not received enough attention because it is still labor-intensive and expensive.

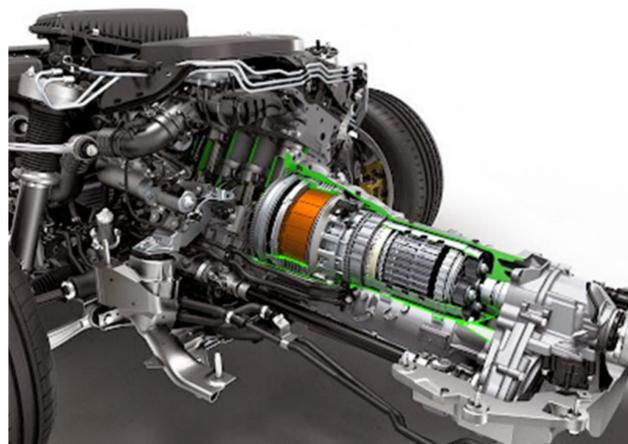


Figure 19. Powertrain of 2022 BMW X5 PHEV [57].

Figure 20 is a schematic of the current automotive scrap shredding and separation methods. It includes processes such as magnetic and eddy current separation, flotation, electrostatic separation, etc. The complexity of the current process would limit the recovery of REE in the Nd-Fe-B content of the ferrous stream. The black box with a question mark in Figure 17 represents the lack of processes that can enable the recovery of the Nd-Fe-B component of the scrap ferrous materials' stream for further processing to REE compounds.

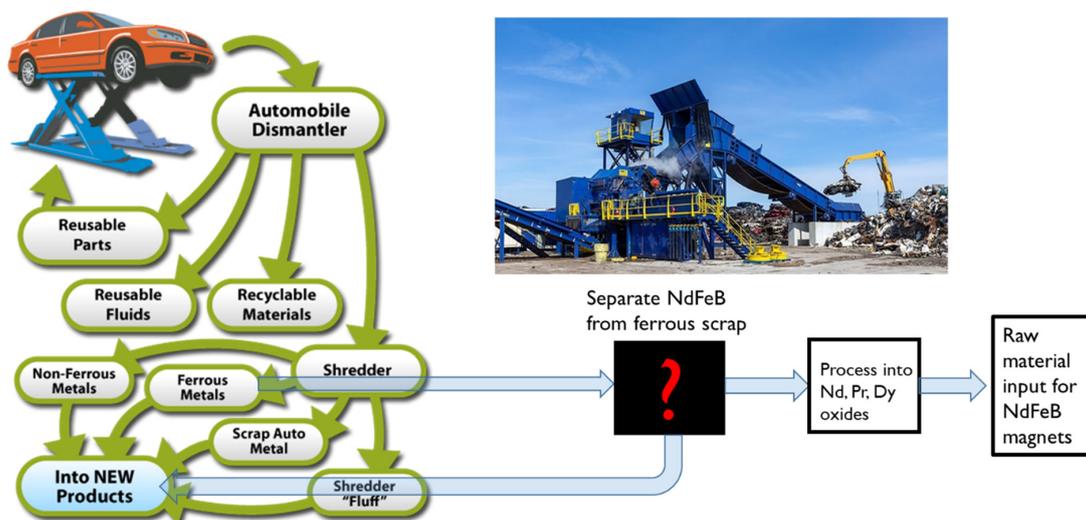


Figure 20. Typical automotive shredding and recovery process.

5. Recycling of Permanent Magnets

Significant amounts of scrap magnets in magnet manufacturing plants are recycled as part of subsequent production steps. However, grinding swarf generated during the machining of permanent magnets are typically contaminated with cutting media and fluids, as well as being oxidized. Such materials are generally suitable for recycling approaches that enable the recovery of the constituent REEs, typically as REOs, that will be subsequently reinserted in the supply chain.

Since the permanent magnets would eventually be used in applications, they mostly reach the point of being recycled when the devices and systems that contain them reach their end-of-life. Thus, the magnets eventually become part of a wider collection of waste, commonly referred to as electronic waste, or e-waste. This presents some challenges, depending on how the different types of e-waste are decommissioned. It also presents opportunities to establish e-waste as a clean energy resource. These challenges and opportunities are further discussed below.

5.1. Challenges and Opportunities in Recycling of REEs from E-Waste

Mitigating the negative health and environmental impacts of the toxic chemical contents of e-waste due to improper disposal is yet another challenge. Challenge and opportunity can be considered two sides of the same coin. For example, the REEs obtained from mining are heavily diluted in concentration, whereas the materials coming from e-waste, such as HDDs and EVs, were previously refined, and hence may require fewer processing steps.

(i) Collection

The challenges in collecting e-waste for value recovery remain among the greatest primary bottlenecks. For example, while an estimated 53.6 metric tons (Mt) of e-waste was produced globally (7.3 kg per capita) in 2019, only 9.3 Mt (~17%) of the e-waste was documented as properly recycled in the same year. That leaves nearly 44.3 Mt of undocumented e-waste that might have been land-filled. The land-filled e-waste was valued at USD 47.6B in 2019, which is expected to increase in the years to come. The global e-waste generation is projected to rise to 74.7 Mt in 2030 and to 110 Mt in 2050 (Figure 21) [58–60].

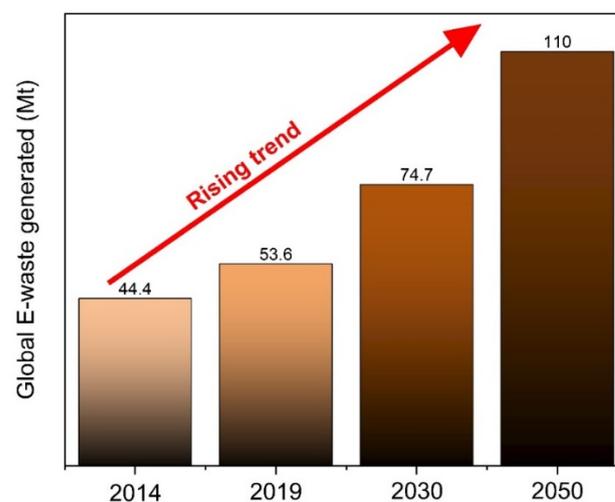


Figure 21. The rising projection in e-waste over the coming years. The plot was made by extracting data from [59].

In addition to the loss of embodied value, if e-waste constituents are not properly recycled, they pose different levels of threats, owing but not limited to toxic elements such as lead, mercury, cadmium, manganese, nickel, and chromium. Other toxic organic

compounds include, but are not limited to, polychlorinated bisphenols, flame retardants, perchlorates, thiocyanates, phthalates, bisphenols, and organophosphates [61]. Improper disposal of e-waste could also lead to emissions of 98 Mt of CO₂ equivalent, 71 kt of brominated flame retardants, and 50 t of mercury emissions by 2050. Consequently, a proper e-waste collection mechanism is needed to ensure that there is little to no contamination of the environment. Most of the recycling needs to be better managed, as can be observed from the waste generated and the waste managed appropriately for the different continents (Figure 22) [59]. Another challenge is the availability of e-waste and the diversity in its distribution across various states in one country. For example, in 2019, the EV waste generated in California was nearly half of that of the other combined 50 states of the US. Of those, 24 states had less than 1% EV waste produced (Figure 23) [62].

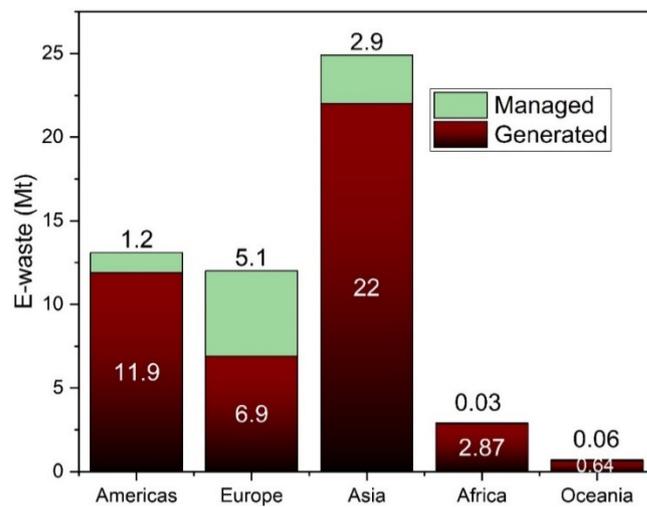


Figure 22. Statistics of e-waste production and its sound management in the different continents in 2019. The plot was made by extracting data from [59].

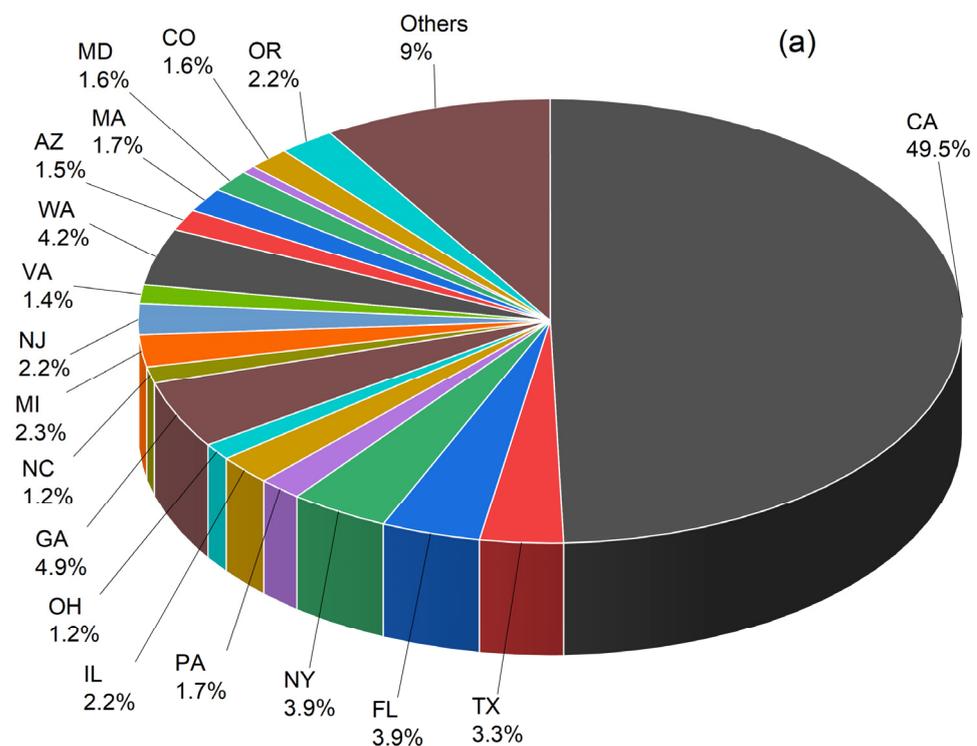


Figure 23. Cont.

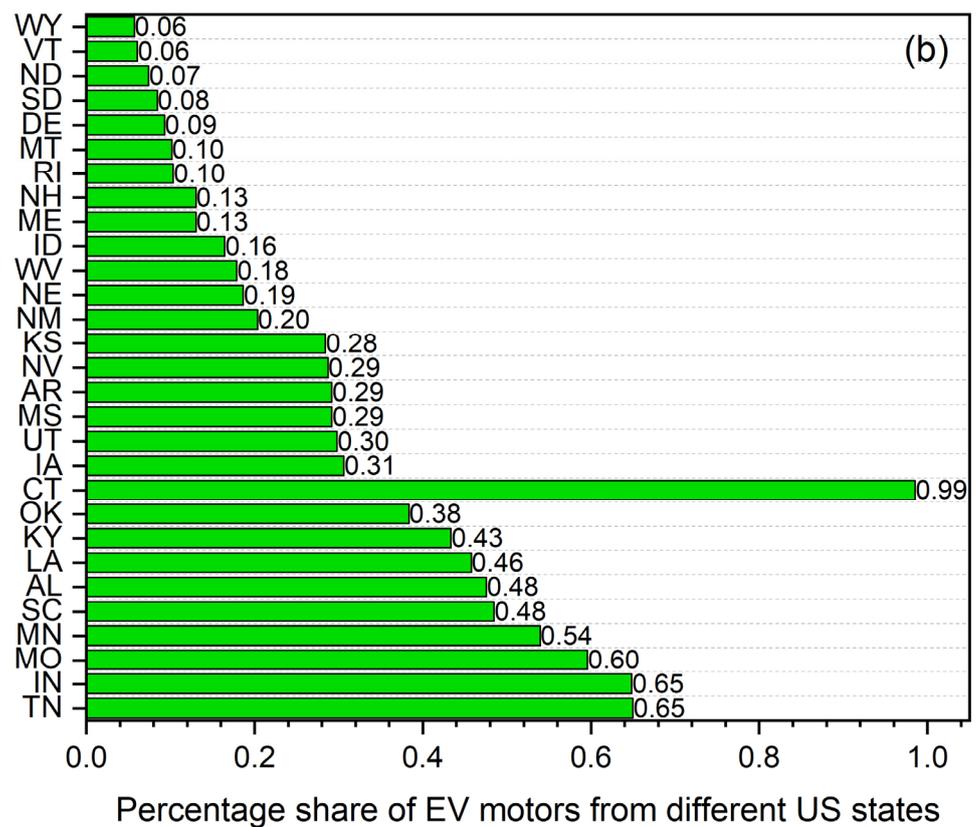


Figure 23. Collection of EV motors from different states of the United States of America in 2019. Data adapted from [62]. The pie data (a) shows the collection of EV motors from all states of USA. (b) The percentage share of states having less than 1% [grouped into others in (a)].

The situation with waste HDDs is more organized across the various states of the US: 19 of the 50 states had less than 1% of HDD waste produced, while California produced a maximum share of 12.3% in 2019 [62] (Figure 24). However, the global demand in recent years has decreased due to solid-state drives (SSDs), as shown in Figure 25a. The HDD demand for the USA is computed based on the ratio of the world and USA GDPs and the global HDD demand in [63].

Two applications, namely, e-mobility and wind turbines, would have the highest recycling potentials, which show sharp growth after 2030 [58,59]. In line with achieving the zero-emission target in the transportation sector, EV sales have been rapidly increasing globally in recent years (Figure 25b) [64]. A demand forecast for EV units in the US shows a rapid increase: 4.72 million unit sales by 2030 [65]. Permanent magnet motors are the most widely used motors in transportation due to their high efficiency and power density. Therefore, in the future, high demand for REE magnets can be expected for traction motors. Additionally, EVs have a significant number of auxiliary motors along with the main traction motors, and some of these motors might be equipped with Nd-Fe-B magnets with various magnet masses. An average magnet mass of 175 g/vehicle was assumed to consider these auxiliary motors in recycling [66,67]. A significant amount of feedstock materials for recycling from EVs will become available in 10–12 years [68]. In addition, the infrastructure for collecting, transporting, and subsequent value recovery from the EOL EVs need to be established. Moreover, the processing, including the removal of magnets from the EOL EVs, is also a complicated procedure, as manual disassembly can be too expensive and a time-consuming process [69,70].

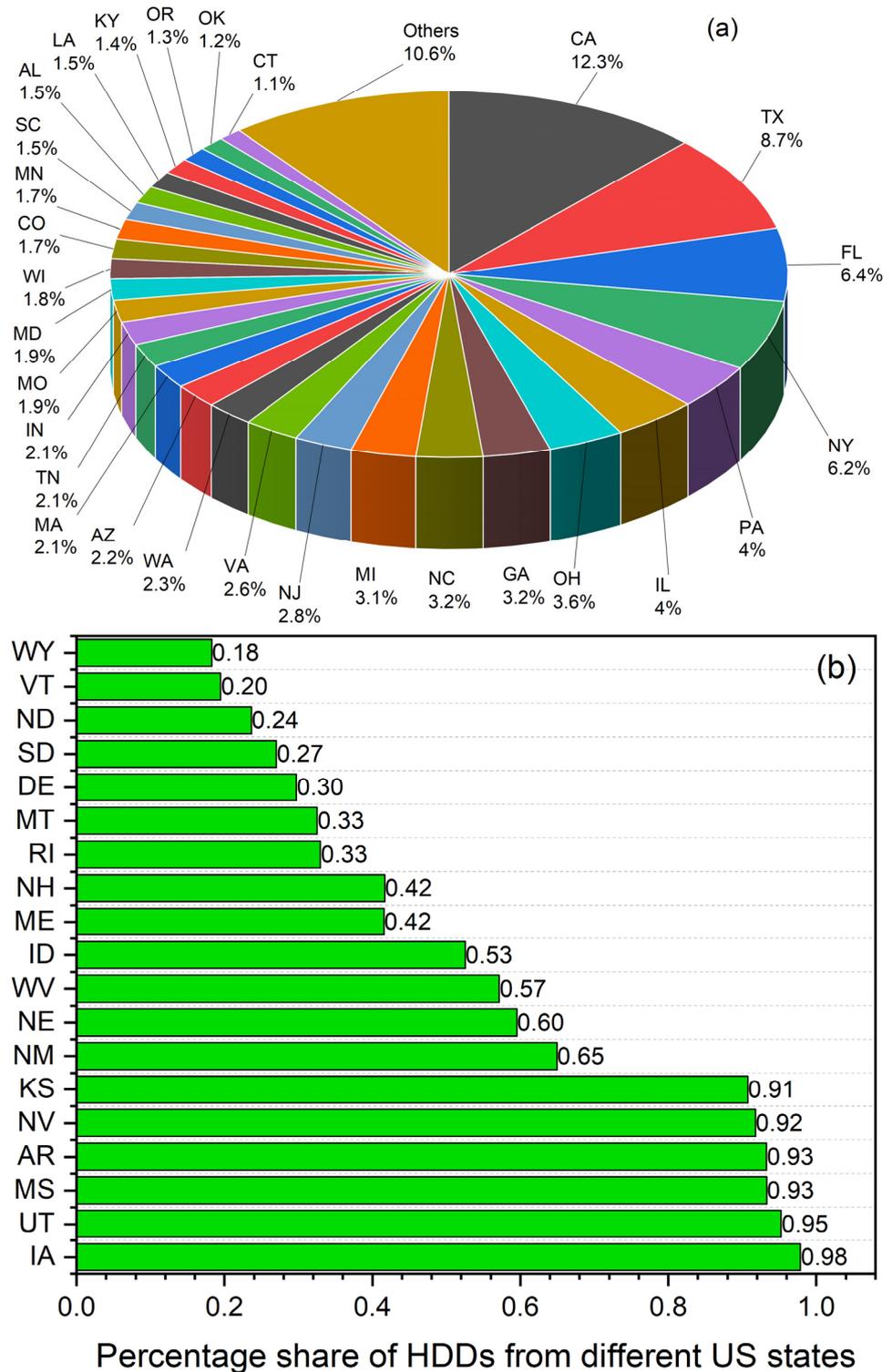


Figure 24. (a) Collection of HDDs from different states of the United States of America in 2019. Data adapted from [62]. The pie data (a) shows the collection of HDDs from all states of USA. (b) The percentage share of states having less than 1% [grouped into others in (a)].

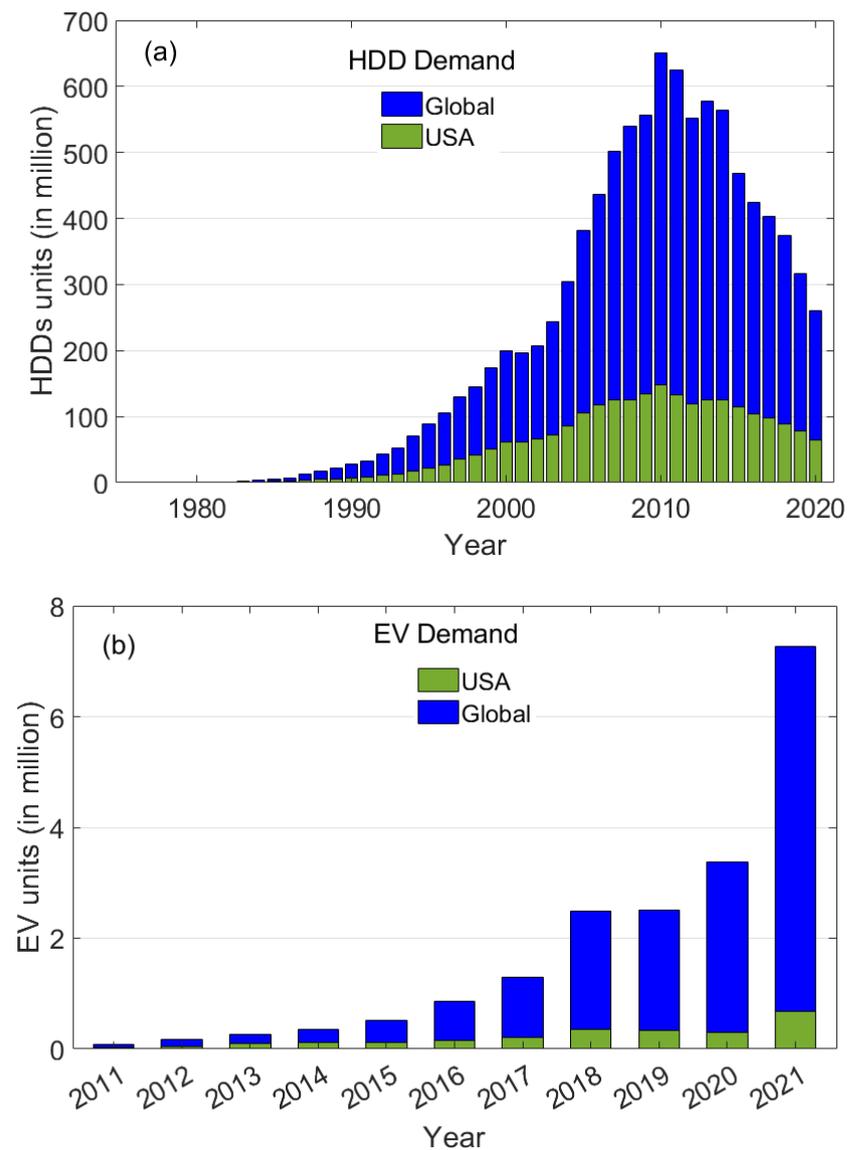


Figure 25. Global/USA demands for (a) HDDs and (b) EVs.

In the USA, only 10.3% of the total electric energy came from wind energy in 2022 (Figure 26a), most of which was from the onshore plants, which generally use non-PM generators [71]. However, offshore wind turbines with a power rating of more than 10 MW are considered cost-effective by improving installation cost per kW. Therefore, to reduce the weight and size of the offshore wind generators, rare-earth permanent magnets are used. Moreover, offshore plants also use the more reliable direct-drive technology to avoid induction generations with gearboxes that are prone to failure [72].

Currently, the USA has two operating plants: the Block Island Wind (50 MW) and the coastal Virginia offshore plant (12 MW). However, recently, the US Department of Energy reported an ambitious target to achieve 22 GW by 2030, followed by 86 GW by 2050 [73]. We project that the demand for REE magnets, especially Nd-Fe-B magnets, will increase up to 675 Mt/year until 2050 for this offshore plant expansion in the USA. This demand also includes magnets for expansion and replacement. Materials for recycling can be expected after 2034, as some of the early installed turbines will reach their EOL [74].

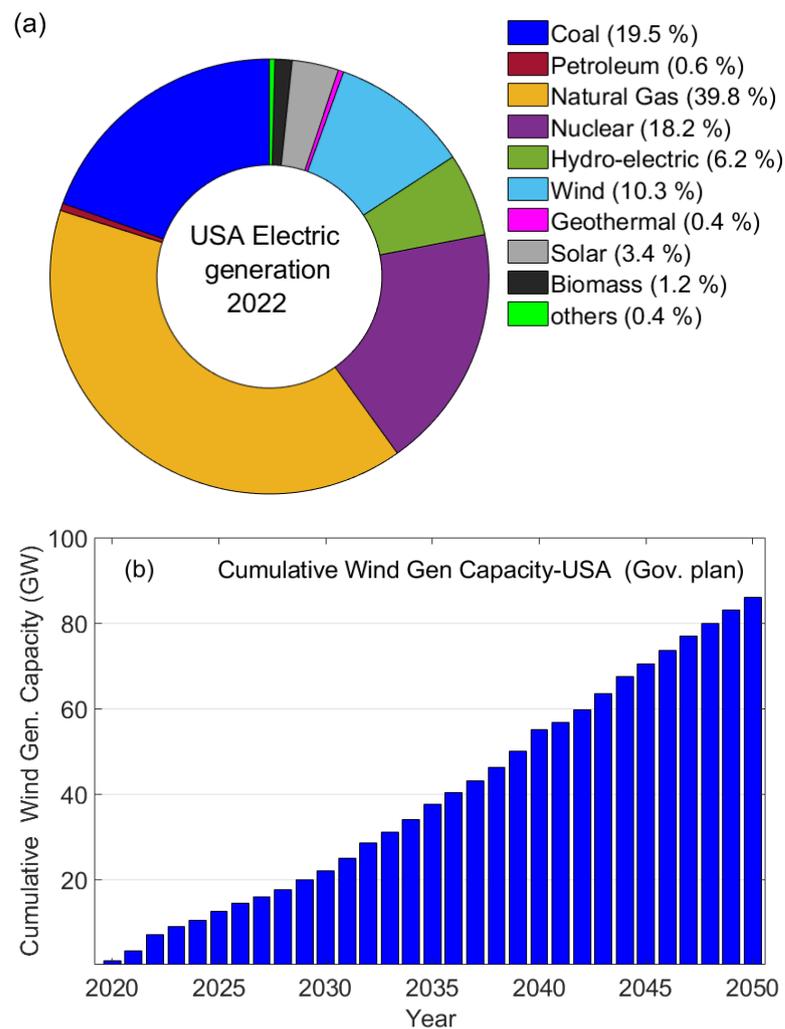


Figure 26. (a) Contribution from different sources for the USA electric generation in 2022. (b) Planned offshore plant generation capacity in the USA. Data adapted from [72,74].

Another challenge is the huge disparity in the weight of magnets used in different applications. Barring four applications, namely, electric vehicles, conventional vehicles, and onshore and offshore wind turbines, there is less than 0.35 kg of magnet usage per appliance in most other applications (Figure 27). Hence, only 30% of the magnet weight comprises REEs, i.e., only 100 mg of REEs is used in most applications. The situation is aggravated further by the fact that the four applications that can yield the largest stockpile of waste magnets are also the ones that require a longer time (>20 years) prior to being available for REE recovery (Figure 28) [75].

Figure 29 demonstrates the visual depiction of the usage of magnets in various applications and the vast disparity in the usage. For example, magnets obtained from three HDDs are equivalent to that obtained from one refrigerator, and magnets obtained from four electric bikes are equivalent to that obtained from one electric vehicle. In contrast, magnets obtained from four hundred electric vehicles can make up for one magnet of the wind turbine, and ten such magnets from wind turbines can make up the magnets required in a magnetic resonance imaging (MRI) unit (Figure 29) [76].

There are a few big projects that have investigated pre-processing of e-waste for REE recovery. For example, the VALOMAG project, a strategy for recovering REE from HDDs, was developed in Europe. To make the process sustainable and economically feasible, the project adopted an automation strategy at every step (Figure 30) [77]. The United States Department of Energy's Critical Materials Institute team of researchers from Oak Ridge

National Laboratory developed a process via which magnets, circuit boards, and other valuable materials can be disassembled using automated technology. This process opens two possibilities: direct reuse of the magnets in applications and/or recovery of critical REE materials from the magnet [78].

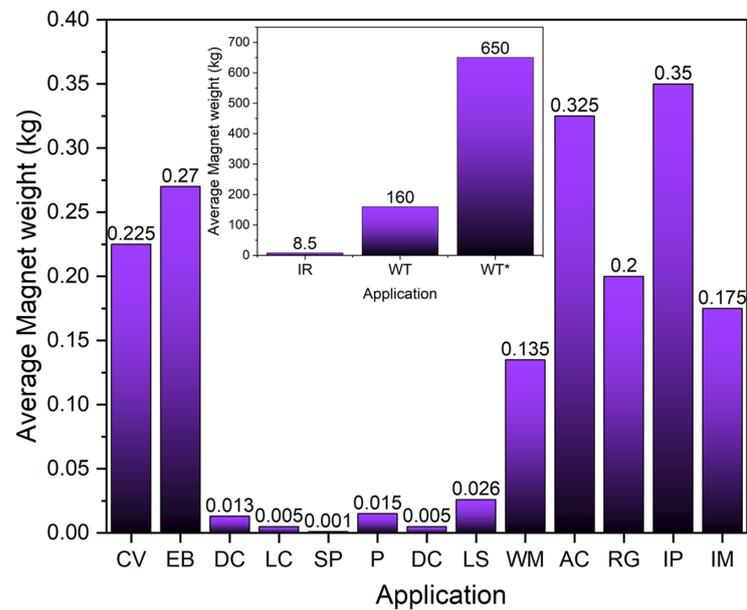


Figure 27. Average magnet weight in different applications. The plot was made by extracting data from [75]. CV: conventional vehicle, EB: electric bike, DC: desktop computer, LC: laptop computer, SP: smartphone, P: printer, DC: digital camera, LS: loudspeaker, WM: washing machine and dryer, AC: airconditioner, RG: refrigerator, IP: industrial pump, EV: electric vehicle, IR: industrial robot, WT: wind turbine (onshore), WT*: wind turbine (offshore).

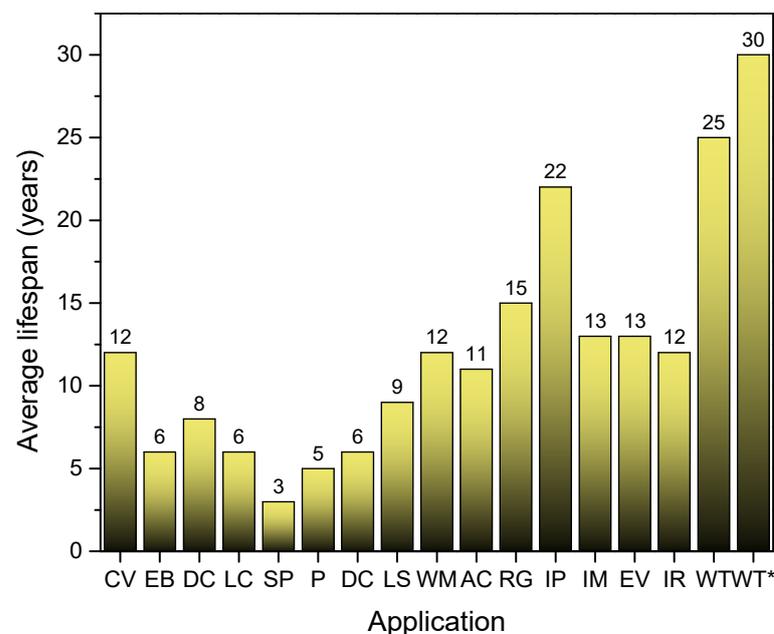


Figure 28. Average magnet lifespan in different applications. The plot was made by extracting data from [75]. CV: conventional vehicle, EB: electric bike, DC: desktop computer, LC: laptop computer, SP: smartphone, P: printer, DC: digital camera, LS: loudspeaker, WM: washing machine and dryer, AC: airconditioner, RG: refrigerator, IP: industrial pump, EV: electric vehicle, IR: industrial robot, WT: wind turbine (onshore), WT*: wind turbine (offshore).

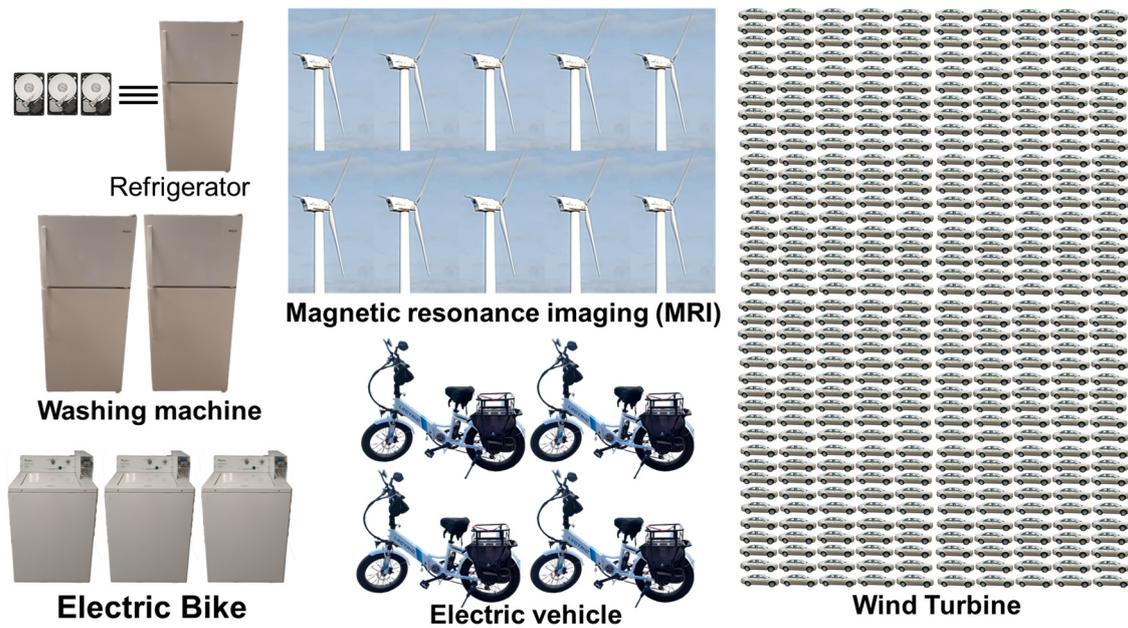


Figure 29. Visual depiction of magnet usages and their proportions in different applications. The plot was made by extracting data from [76].

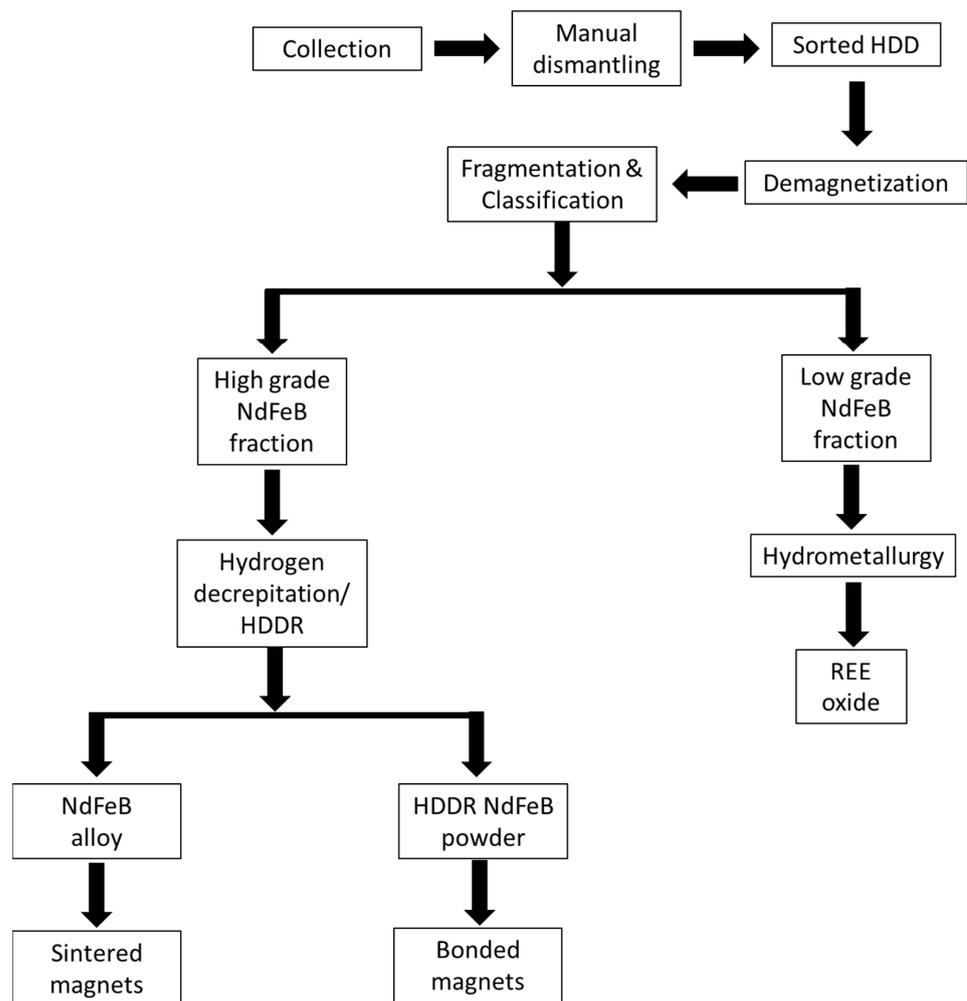


Figure 30. Proposed material reuse and recovery in the VALOMAG project. Adapted from [77].

(ii) Pre-processing

Pre-processing refers to the important prerequisites before a permanent magnet is recovered from the e-waste for direct reuse, indirect reuse, or recovery. Different e-waste would require different pre-processing techniques because the size and mass of permanent magnets vary from each source. Additionally, the design of the systems housing the permanent magnets varies. For example, unlike hard disk drives, permanent magnets recycled from electric vehicles and wind turbines would require different approaches. While the former is preferably shredded, and hence typically requires further processing prior to REEs' recovery, the latter may require disassembly to extract the permanent magnets, thereby enhancing the possibilities of direct reuse and reducing contamination of the magnets.

Li et al. suggest that the recycling of EVs in the future should advance from a one-stage approach to a three-stage approach to decrease the contamination of the valuable materials that can be obtained (Figure 31). In the one-stage approach, the EVs are sent directly to the production line to separate and shred valuable materials, followed by recovery. In the three-stage approach, they propose that the EVs would at first be disassembled into the components, and these would further be segregated into sub-assemblies, followed by shredding and recovery to decrease material contamination [79].

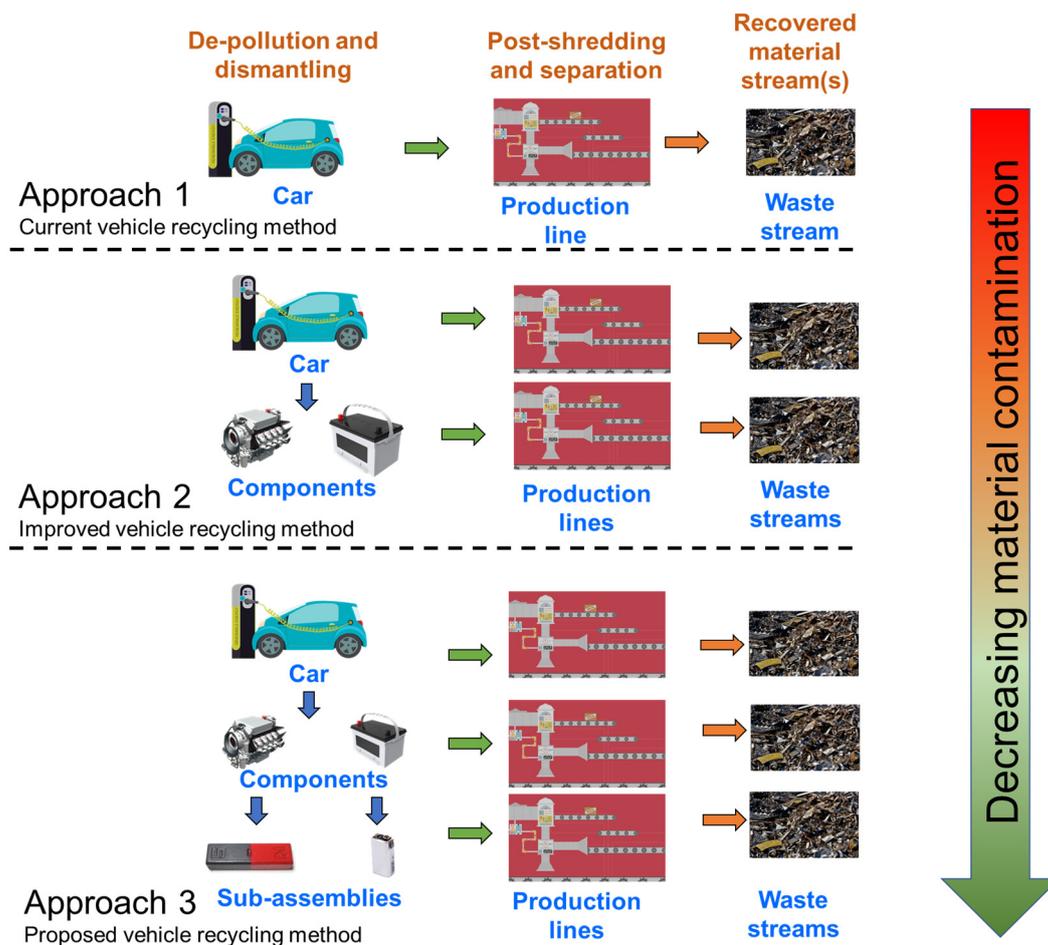


Figure 31. Proposed robotic disassembly approaches to accelerate recycling and minimize critical material contamination. Adapted from [79].

5.2. Recycling of Magnets—Terminologies

There are various terminologies and conventions used to define recycling processes. There still needs to be a clearer definition, as at present, it varies from one publication to the next. For example, Li et al. have referred to magnets cut into small pieces and used again

in a different application as direct reuse [80]. Gandha et al. have cryogenically crushed AM-generated polymer-bonded magnet waste and reused the powder to produce bonded magnets. The authors state that the same process can be applied to EOL-bonded magnets and referred to this process as recycling [81]. It is worth noting that the actual magnetic property, which is restricted to the individual particle, remains intact even after cryogenic crushing. Although one might argue that the property might be much lower than sintered magnets, the crushed powder still possess hard magnetic properties typical of permanent magnets. Thus, the question that arises is, what constitutes a direct reuse and what should the converse be called? In the present report, we categorize the entire recycling into two major divisions, namely, direct recycling and extended recycling (Figure 32). Instead of referring to the converse of direct recycling as indirect recycling, we have chosen extended recycling to avoid confusion. The direct recycling would include minimal damage to the original form of the magnets, although the definition of “minimal” is still subjective. Extended recycling, on the other hand, would include processes that prolong the reinsertion of the magnet or values recovered from them into an application. These processes typically require additional changes to the original form of the magnets, i.e., crushing, grinding, dissolution (hydrometallurgical), melting (pyrometallurgical), etc. Direct and extended recycling will be further discussed in the subsequent sections.

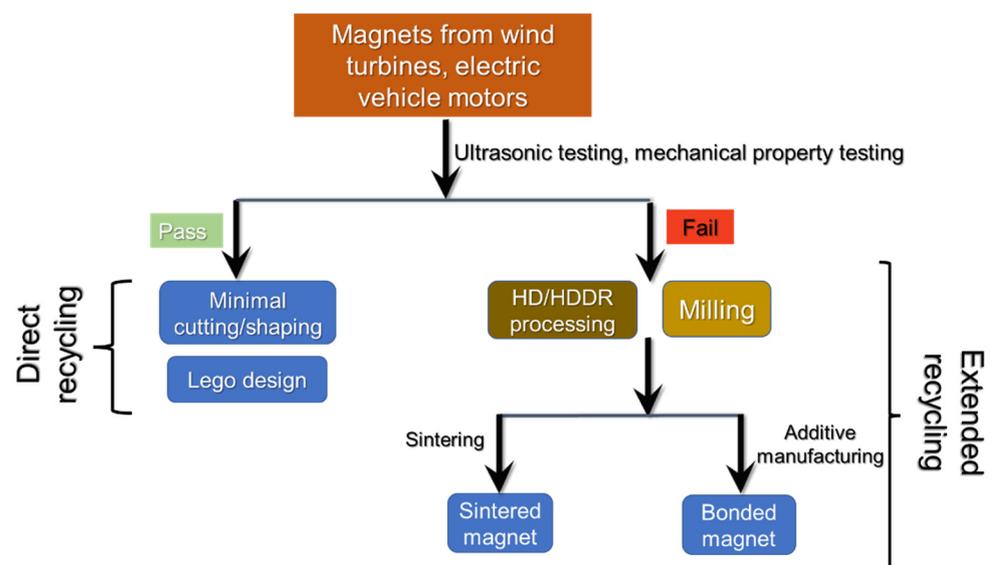


Figure 32. Schematic of direct and extended recycling of end-of-life magnets.

5.3. Direct Recycling and Extended Recycling

Lansink’s ladder can be used to assess the life cycle of any material. It comprises five major steps: prevention, reuse, recycling, recovery, and disposal. The shape of the inverted pyramid can be used to comprehend the importance of each step in the end-of-life value of rare-earth materials. It can also be used to gauge the extent of environmental degradation, highest at the top, and lowest at the bottom. We can expand the hierarchy in terms of magnet recycling strategies. The first step of the ladder represents prevention (Figure 33). Harper et al. describes this as eliminating the use of critical elements [80]. The key approach for prevention is the development of substitutes with no or fewer critical materials for the same applications. Attempts have been reported on the reduced use of critical REE materials in PM motors and generators, which are used for traction/generation purposes [82–86]. Solid-state drives (SSDs) are now replacing HDDs and they do not have critical REE permanent magnets. Therefore, it can be expected that HDD sales will decline due to the new SSD technology [63]. Nevertheless, this trend may be limited by advancements in HDDs with the same speed as SSDs, since HDDs offer lower storage costs. Such an advancement was recently announced by Seagate [87]. Moreover, research is

ongoing for high-temperature superconducting (HTS) generators for wind turbines [88]. Reuse is the next most viable option if environmental impacts are considered.

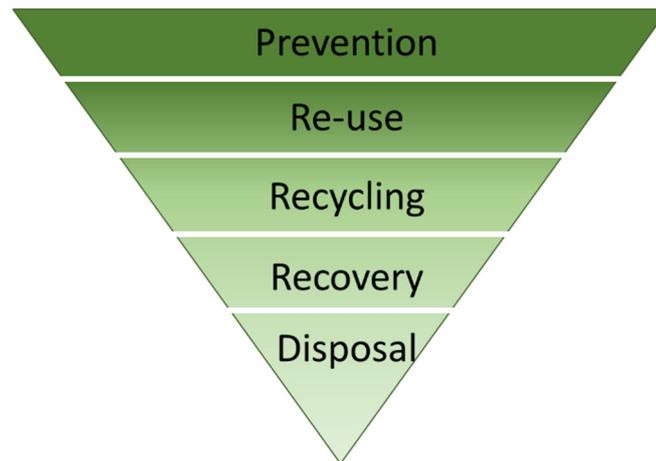


Figure 33. Waste management hierarchy utilized in magnet recycling. Adapted from [89].

The direct recycling of REEs in permanent magnets can reduce the cost, time, and energy of reprocessing used magnets by dissolution, metallization, and magnet fabrication. Hogberg et al. have demonstrated that segmented poles can substitute a bread-loafed-based solid-pole magnet in a direct-drive wind turbine generator or electric vehicle motor [90]. It was demonstrated that using such segmented poles, referred to as the Lego design, instead of a single piece of magnet, a reduction in Eddy current losses was achieved, which helped in achieving comparable values of torque, cogging torque, and torque ripple [90]. The Lego design can efficiently aid in future usage of these segmented magnets, based on the shape and size requirements (Figure 34) [80]. The rotor is subjected to heating in a furnace to easily separate the demagnetized magnets from the rotor core, and a shield can be used to avoid any damage to the furnace or magnets. Magnet coatings with epoxy or metals, such as Cu, Ni, and Zn, also affect the magnetic properties in the thermal demagnetization process. Thus, this process can be used for extracting and reusing permanent magnets obtained from motors [91]. The process of segmenting the magnets will generate some waste that will need to be recycled.

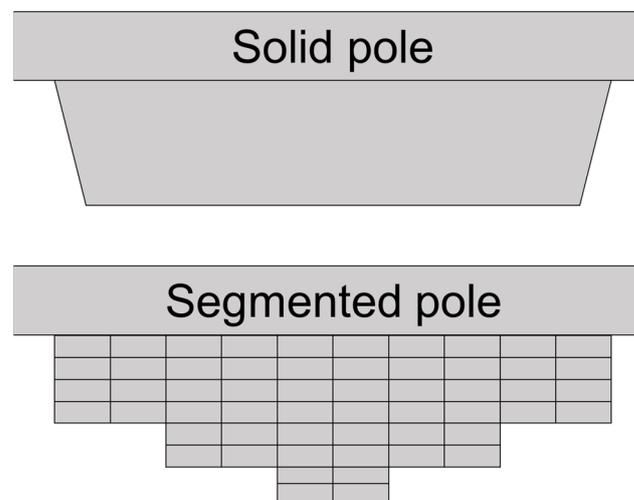


Figure 34. Adopting the Lego design to produce a segmented pole to replace a solid pole: an example of direct reuse. Adapted from [80].

One of the biggest challenges in the direct recycling of magnets is the ability to qualify the magnets as defect-free. Cui et al. have demonstrated the usage of ultrasonic testing to detect the possible presence of internal defects, such as microcracks, porosities, and inclusions [92]. This technique can predominantly aid manufacturers in better understanding the conditions that lead to defect-free samples. Additionally, this method can also help in the identification of end-of-life magnets that can be directly recycled to new magnets.

Magnets that fail to be directly recycled due to poor mechanical properties can be converted to powder and subjected to extended recycling. The usage of hydrogen as a tool to aid in the conversion of sintered magnets to a fine powder with a much finer grain size has gained momentum over the last two decades. The two methods include hydrogen decrepitation (HD) and hydrogenation disproportionation desorption recombination (HDDR) processes [93,94]. Reports suggest that the extended recycling of the fine powders obtained from magnets that were subjected to the HD and HDDR processes, followed by sintering, has helped achieve comparable results to sintered magnets prepared from virgin resources [95,96]. The fine powder obtained using HDDR processing can also be used for bonded magnets by combining with a polymer. The particle morphology, size, ratio of polymer to magnetic material, etc., play a role in the performance of bonded magnets [97]. Gandha et al. have demonstrated that the end-of-life-bonded magnets can be reprocessed using cryo-milling followed by isostatic pressing to achieve energy density values similar to those of starting additively manufactured bonded magnets (Figure 35) [81]. There still needs to be clarity about when a magnet can be sent for reprocessing by HD/HDDR and re-sintered or made into a bonded magnet and when it should be sent for the recovery of constituent REEs.

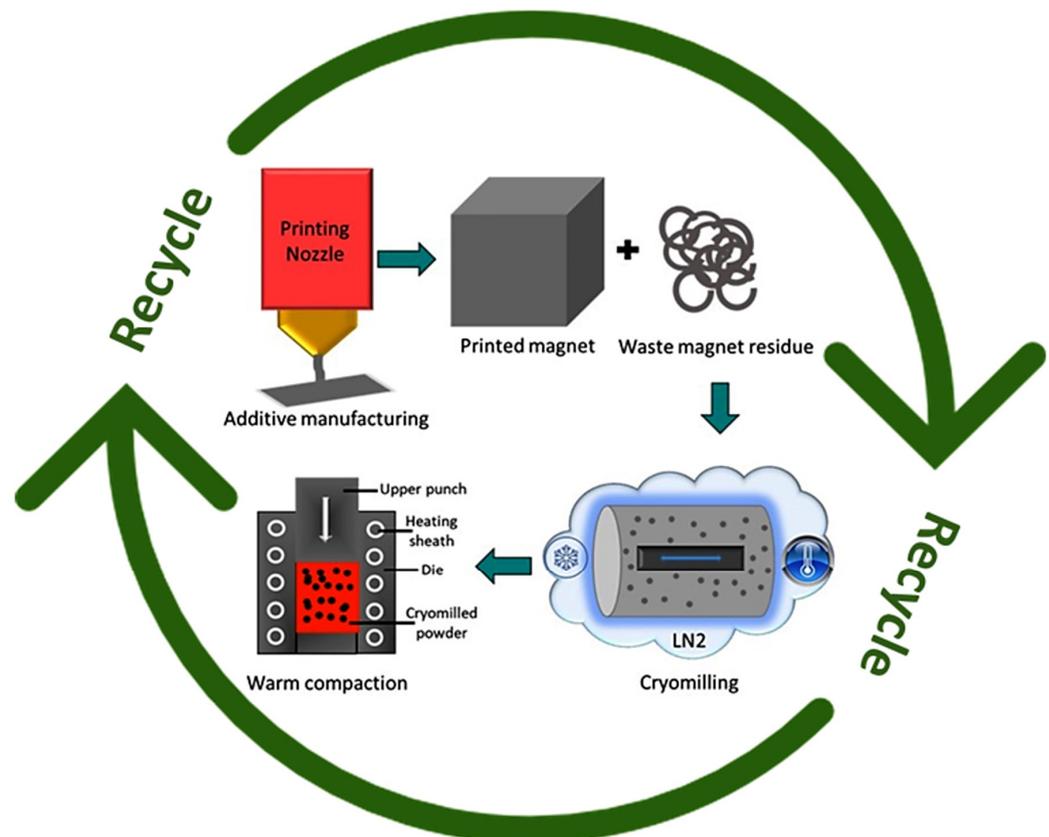


Figure 35. Schematic of the indirect reuse of bonded magnets. Reprinted (adapted) with permission from [81]. Copyright 2019 Elsevier.

5.4. Recovery—A Subset of Extended Recycling

The suitability of scrap magnets for direct and extended recycling can be assessed based on their conditions. For example, if the scrap magnet is significantly oxidized and mechanically damaged, then the recycling can be performed through the recovery of the REEs. Moreover, the recovery process might be the best option when the magnets are not easy to disassemble, especially when they are destroyed together with the device that housed them in the application, e.g., shredded HDDs. Hydrometallurgical and pyrometallurgical processes are typically applied for the recovery of REEs from magnets.

(i) Hydrometallurgical recycling

In the traditional hydrometallurgical recycling process, the EOL magnets are initially demagnetized and subsequently crushed into a fine powder. The powder is then typically oxidized and dissolved with a mineral acid. At this stage, a minimum amount of iron (which now exists as Fe_2O_3) is dissolved, and the undissolved iron oxide is separated by filtration. To the acidic leachate solution containing RE^{3+} , oxalic acid is added, and the $\text{RE}_2(\text{C}_2\text{O}_4)_3$ residue is filtered and separated. The $\text{RE}_2(\text{C}_2\text{O}_4)_3$ residue is subsequently annealed to obtain RE_2O_3 [98]. A hydrometallurgical process was presented in which processing one metric ton of permanent magnet required 13.6 metric tons of water, 250 kg of HCl, and 257 kg of oxalic acid [99]. The process yields a product in excess of 300 kg of REE oxides based on the effectiveness of the recovery process. The electricity input required to process one metric ton of permanent magnets via an example of the traditional hydrometallurgical process is demonstrated in Figure 36. The oxidation process typically consumes the maximum electricity input of 369 kWh [99].

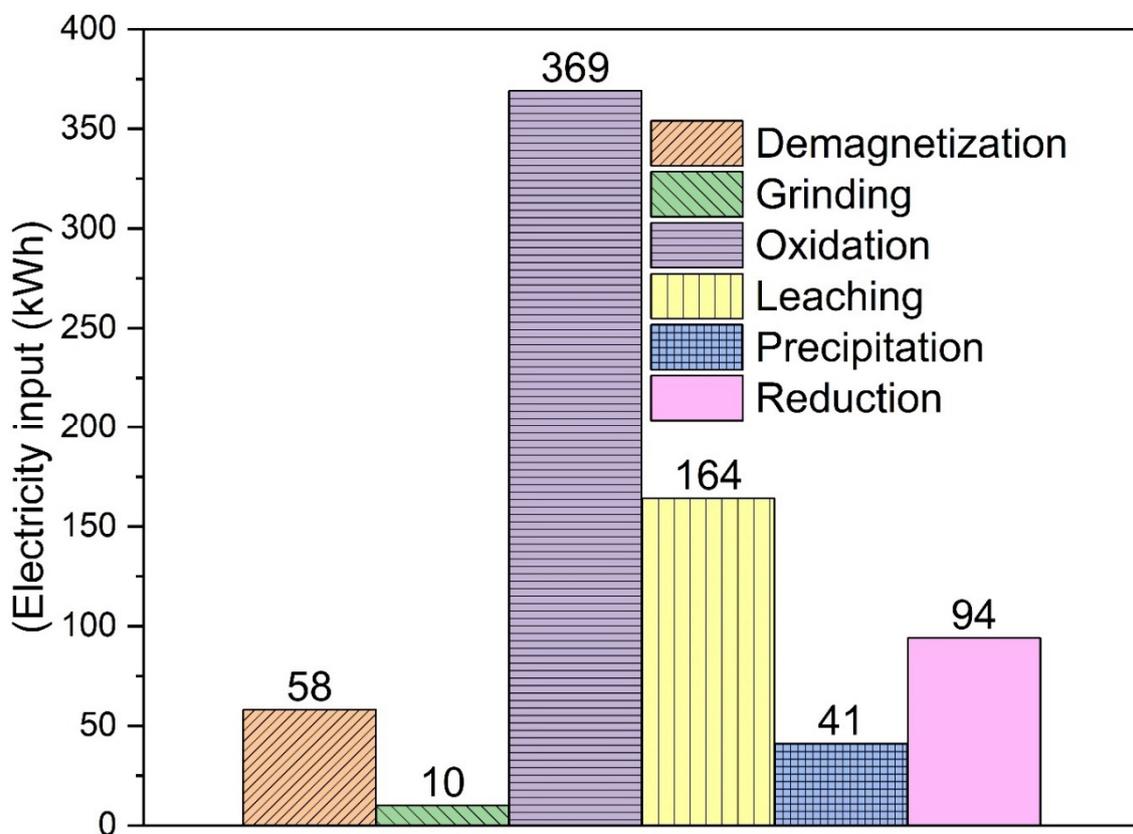


Figure 36. Electricity usage to process one metric ton of permanent magnet using hydrometallurgical processing. The plot has been constructed by adapting data from [99].

In addition to a large amount of acid usage (typically H_2SO_4 , $\text{H}_2\text{C}_2\text{O}_4$, and HF to precipitate the REEs as double sulfates oxalates, or fluorides) and a large amount of

wastewater generation, hydrometallurgical processing typically requires many steps to recover all the REEs [100–103]. Furthermore, separating iron, nickel, and boron to obtain a high-purity RE salt remains challenging. In contrast to Nd-Fe-B magnets, recycling of Sm-Co magnets is simpler. This is because: (a) Sm-Co magnets typically contain only Gd as another REE, thereby decreasing the complexity of separation, and (b) Sm-Co magnets crystallize as 1:5 and 2:17 stoichiometries, where the latter typically contain Fe, Co, Cu, and Zr [104]. We remark that recycling feedstock materials that contain a mixture of Nd-Fe-B and Sm-Co magnets, such as swarf generated during magnet processing, presents elevated-level recycling difficulties. The difficulties owe to the fact that most hydrometallurgical processes co-recover the REEs as a mixture in a product, requiring further separation of the REEs for subsequent permanent magnets' development.

Venkatesan et al. have demonstrated the electrolytic dissolution of the Nd-Fe-B magnet at room temperature using NH_4Cl as the electrolyte. The Nd-Fe-B magnet was used as the anode and a copper rod was used as the cathode. Upon dissolution of the Nd-Fe-B, the Fe^{2+} was oxidized to Fe^{3+} at the inert electrode (Figure 37). The RE^{3+} was subsequently separated from a mixture of $\text{RE}(\text{OH})_3$ and $\text{Fe}(\text{OH})_3$ by using 0.14(M) HCl solution. This is a significant improvement owing to the diluted concentration of mineral acid compared to conventional hydrometallurgical processes [105].

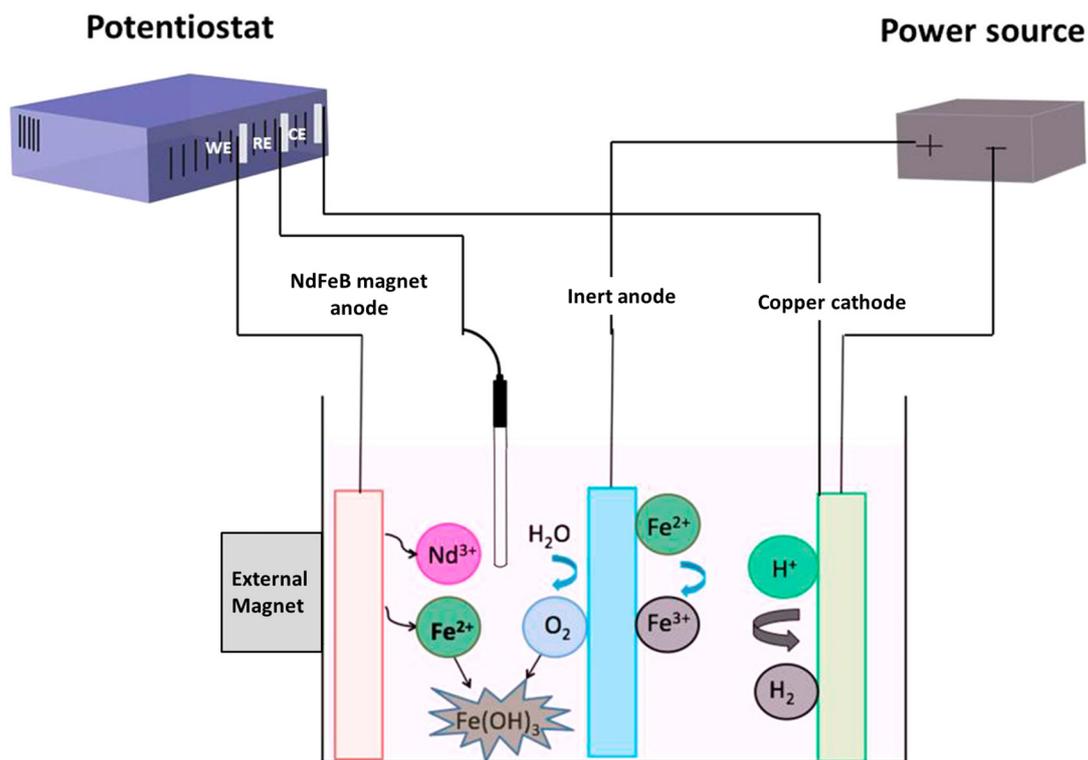


Figure 37. Schematic representation of electrolytic dissolution of Nd-Fe-B magnets. Reprinted (adapted) with permission from [105]. Copyright 2018 American Chemical Society.

There is a concerted effort to depart from the traditional hydrometallurgical routes that require corrosive mineral acids for magnet dissolution. Such efforts in recent research use inorganic salts for the dissolution. Maat et al. have demonstrated the dissolution of Nd from Nd-Fe-B magnets by hydrothermal reaction. The usage of NaCl improves the oxidation capabilities of water. By employing a high temperature (250 °C) and high pressure in an autoclave, atomic hydrogen produced in the process adsorbs on the surface of the magnets and produces a lattice expansion, and thereby, disintegration of the magnets occurs. Subsequently, the Nd converts to $\text{Nd}(\text{OH})_3$, and Fe_3O_4 is magnetically separated.

Furthermore, they demonstrate that the process can also be carried out using other oxidative salts, such as $\text{Na}_2\text{S}_2\text{O}_4$ and $\text{Na}_2\text{S}_2\text{O}_5$ [106].

Prodius et al. have demonstrated the dissolution of Nd-Fe-B obtained from magnet swarf, decrepitated magnets, and shredded e-wastes using CuSO_4 solution at room temperature (Figure 38). Additionally, Sm-Co magnets have also been shown to dissolve via the same procedure. In the presence of aqueous Cu^{2+} and aided by aerial oxidation, the RE^{3+} and Fe^{2+} are converted to $\text{RE}(\text{OH})_3$ and $\text{Fe}(\text{OH})_3$. The Boron is precipitated as copper borate $[\text{Cu}_3(\text{BO}_3)_2]$, and excess copper precipitates as Cu_2O and metallic Cu. The RE^{3+} is extracted as rare-earth oxalate from the leachate. The unique feature of acid-free REE dissolution has brought forth several awards to the technology and the team and has reached pilot-scale commercialization [107–111].

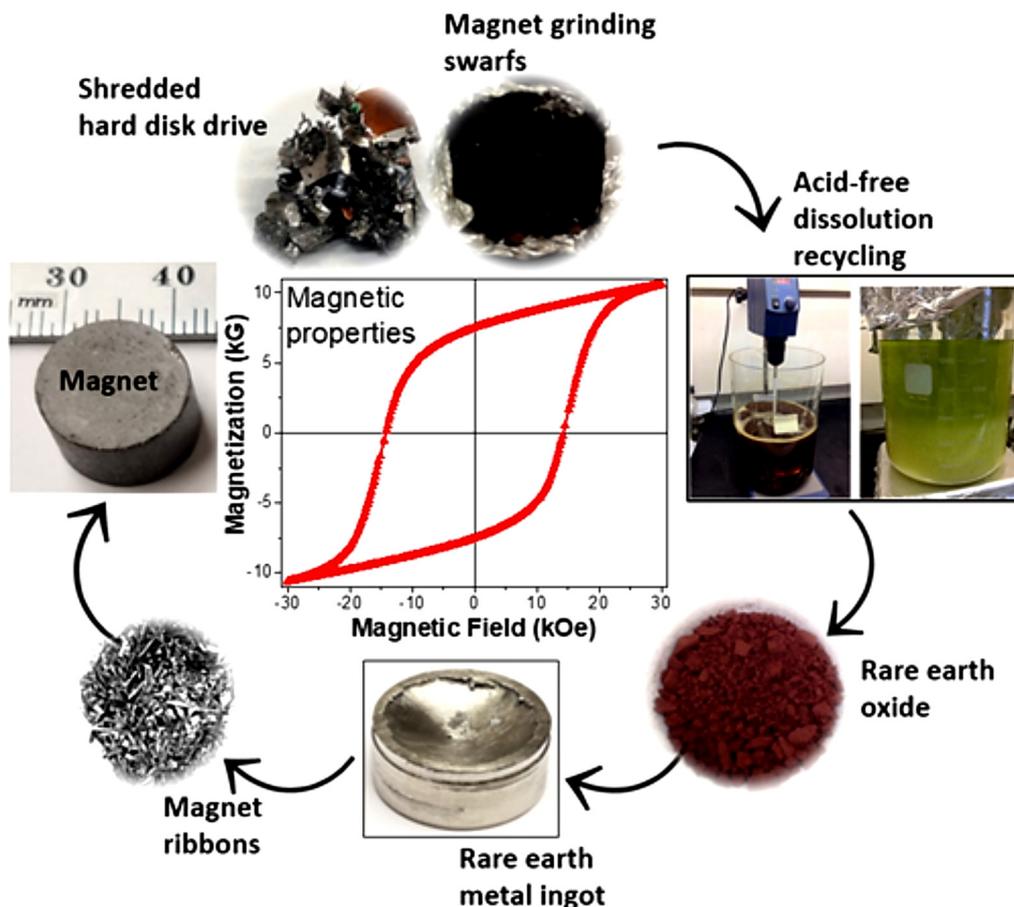


Figure 38. Acid-free dissolution process to make magnets from magnet swarf and shredded HDDs. Reprinted (adapted) with permission from [111]. Copyright 2020 American Chemical Society.

In addition to the dissolution, the separation of rare earth also involves large amounts of chemicals. Furthermore, the process is time-consuming and involves the usage of large separation setups. Prodius et al. demonstrated that using organic salt (typically made by combining an organic base, such as alkyl-imidazolium or alkyl-pyrrolidinium, and an organic acid, such as oxalic acid) dissolved in water to produce neutral pH, the heavy rare-earth oxalates (Gd to Lu) can be separated from the light rare-earth oxalates (La to Sm) in fifteen minutes. The process can be used on feedstock derived from ores and recycling and can also be used to extract heavy rare-earth elements from dilute feedstock (~5%). This opens the possibility of the separation of REEs at neutral pH without the usage of any corrosive mineral acids [112–114].

(ii) Pyrometallurgical recycling

Pyrometallurgical processes do not generate large amounts of contaminated wastewater; however, they require highly corrosive gases, use large amounts of energy, and generate large amounts of solid wastes [115]. In pyrometallurgical processes, liquid metal extraction [116–119] and electroslag refining [120,121] have been predominantly developed and studied [122].

Pyrometallurgical recycling is a very energy-intensive process. For example, Bian et al. state that the reduction step in recovering one metric ton of permanent magnets typically consumes 6688 kWh (Figure 39) and 0.15 kg of argon. The pyrometallurgical recycling of one metric ton of permanent magnets produces 615 kg of Fe and 455 kg of REOs [97].

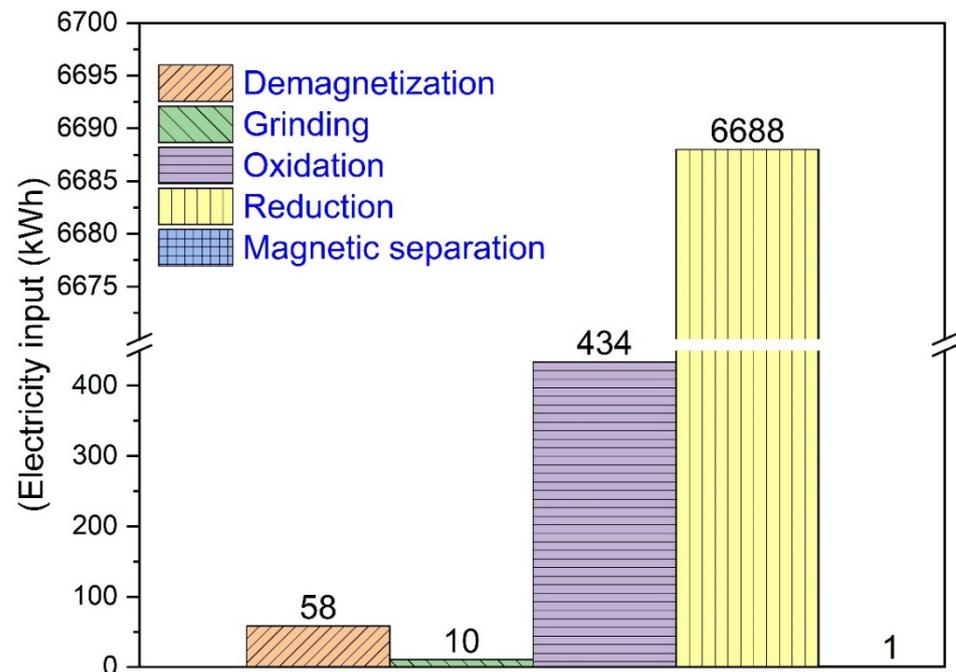


Figure 39. Electricity usage to process one metric ton of permanent magnets using pyrometallurgical processing. The plot has been constructed by adapting data from [99].

In contrast to processes that eliminate toxic and volatile waste products, several reports have been provided on research directions that address this issue using extended processes. Bian et al. have proposed a combination of vacuum induction melting followed by hydrolysis and magnetic separation (VIM-HMS) (Figure 40). In the VIM process, the Nd-Fe-B ingots are mixed with carbon powder and uniformly melted at 1400 °C. Once the melt pool has cooled, they form rare-earth and iron carbides, which are subsequently crushed into a fine powder. The crushed magnet powder is transferred to a flask, and distilled water is added, giving rise to hydrocarbon products in addition to rare-earth hydroxide and Fe₃O₄. The latter product is magnetically separated, which gives rise to 99.7% RE(OH)₃ [123].

Yue et al. have demonstrated the re-fabrication of Nd-Fe-B magnetic powder from magnetic sludge. The sludge is treated with acetone to initially remove the organic impurities. Subsequently, the cleaned and dried sludge material is treated with calcium granules at 1050–1150 °C for 3 h to directly convert the sludge into Nd-Fe-B. The separation of CaO was carried out by dissolution using dilute acetic acid [124].

Saito et al. have demonstrated the complete recovery of Nd from Nd-Fe-B magnets using the flux method. Upon mixing Nd-Fe-B magnets with B₂O₃ and melting the contents, it was demonstrated that complete conversion to Nd₂O₃ was possible. Although the process brings a mandatory separation step of Fe₂B and α-Fe, the process is an alternate pyrometallurgical pathway [125].

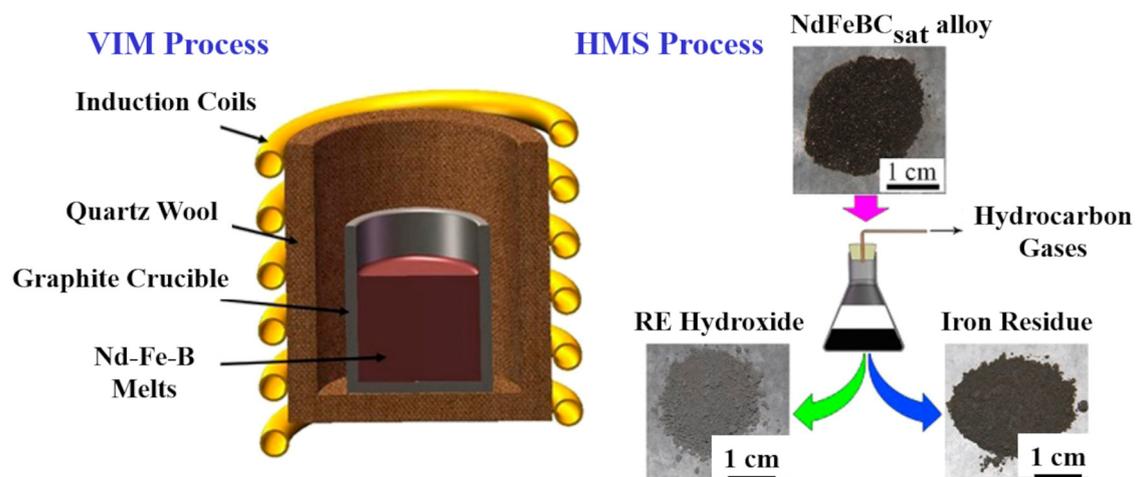


Figure 40. VIM-HMS process to extract RE hydroxides. Reprinted (adapted) with permission from [123]. Copyright 2016 American Chemical Society.

Hua et al. propose the usage of a molten salt mixture of KCl and MgCl_2 to extract Nd from Nd-Fe-B scrap granules. Upon heat-treating the contents, the Nd oxidizes to NdCl_3 , and the boron separates as a solid Fe-B residue. The molten salt mixture is subsequently subjected to electrolysis, which yields the RE-Mg alloy. The proposed technique is an improvement in comparison to both hydrometallurgical as well as pyrometallurgical techniques. The molten salt lowers the initial reaction temperature for the formation of chloride and the electrolysis process ensures that no extra heat needs to be supplied into the system for the separation of the RE alloy. With near complete usage of Fe and REEs, no emission of toxic and corrosive chemicals, and a lower energy consumption, the proposed technology can have an advantage over the existing hydrometallurgical techniques, as well. Another advantage of this process is that it yields REEs in metallic forms, in contrast to oxides that would still require a reduction step [126].

6. Commercialization Efforts

Some efforts are ongoing concerning the commercialization of RE recycling and production to address the criticality issue of REEs present around the globe. One of the earliest commercialization efforts was the partnership developed between Umicore and Rhodia to recycle nickel metal hydride batteries. The team combined the ultra-high-temperature battery recycling process of Umicore and the refining of REEs expertise of Rhodia. A hybrid vehicle NMH battery typically contains about 2 kg of RE, predominantly La, Ce, and Nd. The project aimed to recycle batteries at Umicore, followed by producing REE concentrates in Rhodia's plant in La Rochelle, France [127].

In 2012, AERC Recycling Solutions partnered with Global Tungsten and Powders Corporation to recycle spent fluorescent lamps. They are predominantly rich in yttrium, cerium, lanthanum, and europium, and the project's goal was to recover the REEs from those [128]. Future availability of REEs' recovery from fluorescent lamps will be affected by the significant advancement in the use of LED lamps.

USA Rare Earth LLC teamed up with Texas Mineral Resources Corp. in late 2019 to jointly start a plant in Wheat Ridge, Colorado, to aid in separating and purification of REEs from ores obtained from El Paso, Texas. The plant was their first venture outside China with complete domestication and aiding the US critical materials supply chain from extraction to processing and distribution. The team looked at ion exchange chromatography to process REEs and targeted an annual production of nearly 2313 tons of REO per year [129]. In the spring of 2020, USA Rare Earth LLC purchased the Nd-Fe-B-producing hub in North Carolina from Hitachi Metals America, Ltd. It was proposed that the plant could produce nearly 2000 tons of REE magnets per year [130].

Solvay announced in 2022 that their La Rochelle plant in France would start the manufacturing of separated REOs and proposed to use it for the manufacturing of traction motors, wind turbine generator motors, and other electrical appliances. Traditionally, Solvay had investigated REE separation, purification, recycling, and formulation. The long-term goal of the project was to produce the largest REE hub in Europe [131].

The US Department of Defense awarded USD 35 million to MP Materials for mining and separation of REEs from USA's largest REE reserves in Mountain Pass, California. This would propel the US toward domestic resilience on REEs and thereby decrease their dependency on imports. The REEs separated by MP Materials were planned to be utilized by General Motors to advance their EV ventures [132,133].

Energy Fuels announced the acquisition of seventeen mines in Bahia, Brazil, for extracting Nd, Pr, Dy, and Tb from monazite sands. The province is historically significant for mineral sands containing REE deposits. It was proposed that when fully functional, it can deliver anywhere from 3000 to 10,000 metric tons of monazite concentrate to Energy Fuels. The deal was formally announced in the first quarter of 2023 [134].

A few efforts are looking at recycling end-of-life products to extract critical REEs. For example, the acid-free dissolution recycling of permanent magnets obtained from HDDs and magnet swarm was awarded a patent in 2018 to researchers from the Ames National Laboratory. The technology went commercial with TdVib LLC in 2021 and has received State and Federal funding supports to scale-up REE recycling from e-wastes [110].

In 2022, Noveon Magnetics, a Texas-based US company, announced that they started producing new magnets from end-of-life magnets. According to the company, their process would be 90% more energy-efficient and could reduce the CO₂ emissions by 11 metric tons per ton of magnet production, compared to the production of magnets from virgin sources. Recently, the US DOD awarded them USD 28.8 million for magnet production [135].

7. Future Outlooks

The rising demand for EVs and the subsequent mandates proposed by several countries to achieve net-zero emissions in a certain timeframe warrants the need for research on better recycling and mining strategies. As of November 2022, nearly 140 countries have announced timelines for achieving net-zero emissions. While some countries have already achieved net-zero emissions, several countries are working toward it [136,137].

The announcement by several automotive companies to stop producing gasoline-powered vehicles serves as a major boost to aiding the e-waste supply chain and could open opportunities for research and development on improved EVs, in addition to recycling of the e-wastes subsequently produced. To date, Mercedes [138], General Motors [139], Ford [140], Stellantis [141], Volvo [142], and BMW [143,144] have made announcements on moving away from fuel-based vehicles by 2030–2040.

Global EV sales have been on the rise exponentially over the years, with a major share of this contributed by China and the USA (Figure 41). Among these, the battery-operated electric vehicles (BEVs) are projected to take up a major share. It is estimated that from 9 million BEV sales in 2022, the number is projected to rise to 52 million by 2030 [145].

In the drive for clean energy, the role of wind turbines is going to be decisive in several countries around the world. Most wind turbines are currently based on double-fed induction generators (DFIGs), instead of the more efficient direct-drive permanent magnet generators (DDPMGs). Although the initial investment cost of the DFIG is low, the use of coils in its rotor, compared to permanent magnets in DDPMGs, exposes DFIGs to several disadvantages. It limits their ability to connect and support power grids. They can disconnect from the grid during under-voltage grid fault conditions, which can also take other DFIGs offline, hence leading to periods of inoperability. Moreover, the DFIG is connected to the grid using a back-to-back converter that can convert only 25–30% of the power produced by the DFIG, which is very inefficient in comparison to DDPMGs, capable of full power conversion. These are a few of the disadvantages of DFIGs, which are poised to exacerbate as the turbines age. When the total costs of investments and operations

are considered, DFIGs may become costlier than DDPMGs, even without considering the costs due to the downtimes of DFIGs. The availability of REEs for permanent magnets is a key limitation to transitioning from DFIGs to DDPMGs, and this opens avenues of research opportunities.

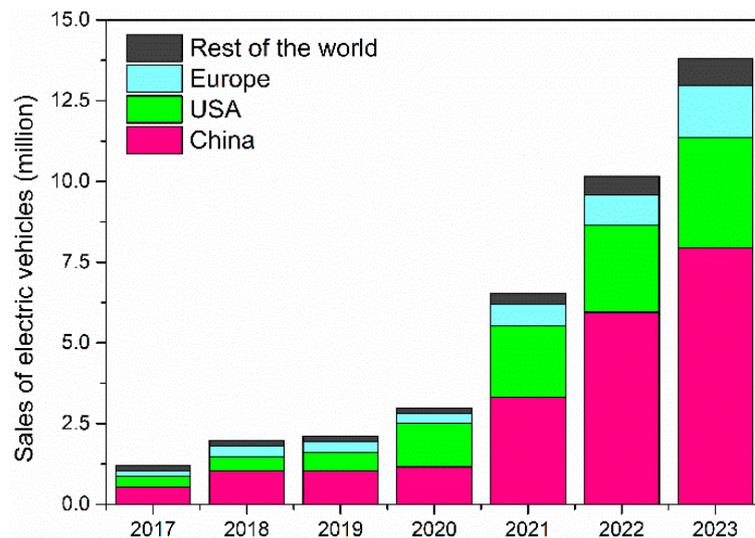


Figure 41. Global EV sales over the years. Replotted by extracting data from [146].

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