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Review

Review of Methods for Obtaining Rare Earth Elements from Recycling and Their Impact on the Environment and Human Health

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Abstract: Rare earth elements (REEs) are at the forefront of discussions, given their crucial role in cutting-edge and eco-friendly innovations that propel the industrial revolution towards a green economy. These elements have become indispensable to various modern technologies, such as smartphones, electronic devices, and renewable energy sources. Many different concepts and analyses have been introduced, such as the chemical similarities among REEs, health risks and ecological damages, the negative environmental impacts of current recovery processes, and strategies for advancing REE recovery towards a circular economy. Although these elements have been widely used in various applications over the last 20 years, the literature on these aspects is fragmented and spread across different research areas, shared by multiple branches and application fields. These fields include safety concerns, economic challenges, and technology. Summarizing and classifying this literature is challenging due to its fragmented nature, the variety of topics, and the different approaches used. The quest for cleaner recycling strategies necessitates a comprehensive assessment covering economic, technological, and environmental aspects. The primary goal of this review is to provide a holistic perspective on REEs, with a central focus on their economic, technological, and environmental dimensions, particularly emphasizing reuse, recycling, and occupational safety. The review begins by addressing complexities of REEs, highlighting the associated technologies, environmental concerns, and economic considerations. It further explores the aspects of reuse and recycling of REEs, shedding light on the advantages, drawbacks, hazards, and costs associated with recycling technologies for REE recovery. Additionally, the review summarizes occupational exposure and safety considerations related to REEs.

Keywords: rare earth elements; recycling; reuse; environmental challenges; economic challenges

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Citation: Gkika, D.A.; Chalaris, M.; Kyzas, G.Z. Review of Methods for Obtaining Rare Earth Elements from Recycling and Their Impact on the Environment and Human Health. *Processes* **2024**, *12*, 1235. https://doi.org/10.3390/pr12061235

Academic Editor: Carlos Sierra Fernández

Received: 12 May 2024
Revised: 6 June 2024
Accepted: 14 June 2024
Published: 16 June 2024

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1. Introduction

Rare earth elements (REEs) wield considerable influence over contemporary lifestyles, yet many people remain unaware of the substantial impacts associated with these elements. It is imperative to recognize the significance of this distinct group of metals, REEs, particularly in the context of their pivotal role in advancing modern industry towards a green economy [1]. Defined by the International Union of Pure and Applied Chemistry (IUPAC) as a collection of 17 chemically similar metallic elements [2], REEs encompass 15 lanthanides along with scandium and yttrium [3]. There are three primary categories of rare earth elements (REEs): light, medium, and heavy. Heavy rare earth elements (HREEs) hold greater economic significance compared to other REE groups due to their extensive practical applications. Additionally, HREEs are typically less abundant in REE minerals than light rare earth elements (LREEs) and medium rare earth elements (MREEs), making them relatively uncommon in terms of high relative abundance. Consequently, it is imperative to separate and extract HREEs from LREEs and MREEs to meet their high economic value and satisfy market demand. However, given the scarcity of REEs overall, the recovery

of all REE groups (light, medium, and heavy) is crucial [4]. It is noteworthy that scandium, distinguished by its unique attributes, and the radioactively classified light lanthanide promethium (Pm), are excluded from the LREE/HREE classification [5]. Among these elements, promethium (Pm) stands out as the only rare earth element that does not occur naturally [6].

Specific REEs display higher abundance in comparison to non-REEs; for instance, cerium (Ce) is one of the most abundant elements on Earth [7,8]. In Mengli Xie's study, the average content of heavy rare earth elements (∑HREE) was found to be 24 $mg \cdot kg^{-1}$, which was lower than that of the upper continental crust and the rare earth element abundance of China's continental crust [9]. Hamzah et al. [10], in an X-ray diffraction (XRD) with scanning electron microscopy/energy dispersive X-ray spectroscopy (SEM/EDS) study of samples from tin tailing sands in Kuala Raya Singkep Island (Indonesia), reported an average content of 0.2% for La, 7.8% for Ce, and 2.0% for Th, while identifying monazite and xenotime as the primary carriers of rare earth elements [10]. The distribution and differentiation characteristics of REEs are influenced by soil pH value and organic matter content; LREEs become more enriched than HREEs with increasing altitude in the basin [11]. The background level of REE content in both surface and subsoil waters varies significantly and is primarily dependent on local geology. Unfortunately, maximum acceptable limits for REEs in drinking water are not provided by any international health organization, and there are insufficient data available regarding their toxicity to human health, as mentioned previously [12]. In general, LREEs are more prevalent than HREEs [13].

Despite the name "Rare Earth Elements", they are not as scarce as the term might imply. Contrary to the notion that REEs are exceptionally rare and challenging to locate, the majority of these elements are, in fact, quite abundant in the Earth's crust overall. In this context, the term 'rare' is employed not to signify their scarcity in terms of natural occurrence but rather to indicate that REEs are considered "rare" due to their limited presence in economically viable quantities [14]. However, the challenge lies in the extraction process, as REEs are often intricately bound to other minerals and are found in low concentrations, making them difficult to extract from ores [15]. Typically, REEs are found in mineral deposits, with over 250 mineral classes identified to date, including carbonates, phosphates, silicates, halides, alkalines, and oxides. Among these, monazite, bastnaesite, and xenotime stand out as the most prevalent REE minerals, serving as primary sources for the production of many REEs [16].

Although REEs share comparable configurations, their distinct physicochemical attributes make them indispensable in a diverse range of applications [17]. These attributes arise from the fact that many REEs are frequently the only viable choice without suitable substitutions for various applications [18]. With these remarkable attributes, REEs find applications in advanced technologies, including lasers [19], permanent magnets [20], and energy storage [21], designating them as essential "technological" metals. The strategic significance of REEs supply has increased as a result of the rising global demand for these metals, presenting a potential challenge [22]. Recognized as "critical" elements, REEs carry economic significance with a high supply risk, influenced by geopolitical factors, particularly China's quasi-monopoly on their production [23].

The complexity of the REE issue is underscored by multiple factors. Despite sustainability assessments in previous research [24,25], the scientific literature in this domain remains fragmented, with limited information on REEs, technological challenges, and safety concerns. The study of REEs is evolving into a complex system, exemplified by the hormesis effect observed in REEs, where low doses promote biological growth, while sufficient concentrations can be toxic to humans [26].

Thomsen et al. [27] documented that even at very small doses of 0.2/kg, gadolinium can induce nephropathy in patients with multiple risk factors. Gadolinium was found to persist in human bone tissue following the administration of 0.1 mmol/kg of Omniscan or Prohance a few days prior to surgery in patients undergoing hip replacement [28]. Lanthanum oxyiodide nanosheets combined with a low dose of doxorubicin demonstrated

increased efficacy in killing A375 cells compared to doxorubicin alone [29]. Despite the increasing focus on rare earth elements (REEs) in toxicology research in recent years, the hazards they pose to human health remain largely unknown. This is primarily due to limitations such as the synergistic toxic effects of various REEs in real-world environments, inadequate detection indicators, and dynamic metabolic differences among individuals [30]. Industrial workers regularly exposed to REEs have reported respiratory, nervous system, liver, and cardiovascular toxicities [31]. Additionally, the physical and chemical similarities among REEs contribute to the complexity of their determination. While there is a drive for technological advancements in REEs, it is crucial to recognize that technology alone is not the complete solution; factors such as cost and safety must be integral to the evaluation of REEs.

This paper addresses the intricate challenges surrounding REEs, defining three main challenges: economic, technological, and safety concerns related to rare earths. Section 2 presents the interdisciplinary issue of REEs, Section 3 delves into current research on the reuse and recycling of REEs, and Section 4 discusses occupational safety issues.

2. The Interdisciplinary Issue of REEs

The significance of reusing REEs is on the rise, bringing together diverse disciplines under the common theme of REEs. Consequently, there exist distinct gaps stemming from various sources that necessitate specific and targeted measures (Table 1).

Table 1. Overview of perceptions on REEs, encompassing literature viewpoints on technology, economics, and safety.

Author(s)	Disciplines with REEs as Common Denominator	Perception in Literature	Challenges as Quoted in Literature
[15]	Technology	REE chemical similarities	REE separation chemistry is a big challenge.
[32,33]	Technology	As dispersed knowledge	Considerable uncertainty and a lack of understanding regarding the consequences of REEs production.
[3]	Environment	Drastic improvement recycling rates for REEs	REE recycling can address the environmental challenges stemming from mining and processing.
[34]	Environment	Recycling methods entail various environmental repercussions	Recycling necessitates significant energy consumption, the utilization of substantial chemical quantities, and the production of waste chemicals and water.
[15]	Economics	As an economic identification demand	The current reduced prices of these elements hinder the advancement of recycling technologies and their economic viability.
[35]	Economics	As a missing evaluation factor	Economically viable extraction presents unique challenges for each waste stream.
[36]	Safety	Considerable number of radioactive elements in REEs	REE mining poses a serious challenge due to thorium and uranium.
[8]	Safety	As a necessity for safety investments	The urgency for new policies addressing the safety of rare earth element mining activities is heightened by concerns about radioactive waste.
[37]	Safety	As a risk management strategy	Brief inhalation exposure to REEs can cause acute irritant bronchitis, while prolonged exposure may lead to pneumoconiosis and progressive pulmonary fibrosis.

REEs have taken center stage in discussions due to their pivotal role in advancing innovations that are driving the industrial revolution towards a green economy. Research groups worldwide have shown a heightened interest in developing recycling processes for secondary streams [17]. Different methods, such as leaching and pyrometallurgy, are being investigated for the recovery of REEs. Numerous prior studies have reviewed potential

technological paths for reclaiming REEs from wastes and byproducts, identifying REE-containing waste streams and characterizing process flows for recovery [35]. Despite the complexities, certain rare materials exhibit high recycling potential, offering economic benefits [38]. However, the extraction and separation of REEs remain expensive and intricate, presenting numerous environmental challenges [34]. An important environmental concern arises from the mining and beneficiation of REEs, particularly their common association with thorium and uranium. Strict control measures are essential to prevent the dispersal of radioactive elements into the environment [39]. The assessment of the potential and effectiveness of REE recycling, along with considerations of secondary sources and production waste, will remain a focal point, necessitating continual examination for different REEs in diverse contexts. This sustained attention is crucial for evaluating the overall environmental impact of REEs and their contribution to advancing "clean energy" [38]. An analysis reveals that both HREEs and LREEs surpass the critical limits, indicating an increased supply risk.

Hence, despite their prevalence in the earth's crust, numerous REEs face supply risks. The European Commission has included REEs in the European list of critical raw materials due to their economic significance and limited availability. In 2017, the European Commission released a new report to categorize critical elements based on their economic importance and supply risks. Similarly, in 2018, the USA also designated REEs as part of the list of 35 critical minerals [40]. The surge in research efforts focused on recycling technologies for rare earth elements (REEs) was triggered by a crisis in 2010 that significantly disrupted the global supply and demand equilibrium for REEs. Consequently, many countries began investigating different alternatives and strategies to diversify their sources of natural REEs [24]. Additionally, the increasing need for rare earth elements (REEs) in advanced technology sectors and eco-friendly initiatives like electric vehicles, wind turbines, and other sustainable technologies has underscored the importance of focusing more on recycling REEs [41].

Although both categories face this risk, LREEs demonstrate more pronounced economic significance. Graedel's three-dimensional graphic [42], depicted in Figure 1, signals potential supply risk for elements such as Eu, Dy, and Er. This projection relies on their potential supply risk, corresponding environmental implications, and vulnerability to supply limitations [12]. Consequently, the escalating global demand for these elements has sparked significant interest among exploration geochemists and technology developers, primarily due to their pivotal role in green applications like hybrid cars, electric vehicle batteries, and wind turbines, against the backdrop of climate change and global warming [12].

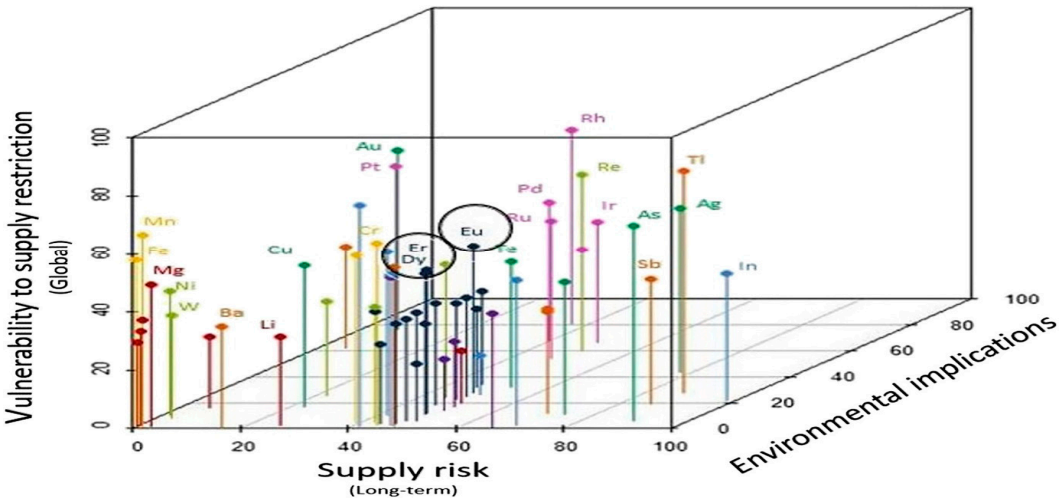


Figure 1. Estimated supply risk for various metals [12].

Researchers have extensively focused on REEs, delving into their economic importance [43–45]. REE-bearing minerals are geographically unevenly distributed worldwide, with the majority of production originating from China (Figure 2), that possesses reserves of monazite. Monazite deposits are also found in several other countries such as Australia, South Africa, Brazil, Malaysia, India, and Russia [46]. Markets and Markets predicts the REEs market growth USD 9.6 billion by 2026 [47], with a 12.3% CAGR from 2021 to 2026. Fortune Business Insights forecasts a growth of USD 5.52 billion by 2028 [48], to rise at a CAGR of 10% during 2021–2028. Global Market Insights estimates the market was USD 13.2 billion in 2019, expecting a CAGR of over 10.7% from 2020 to 2026, reaching USD 19.8 billion by 2026 [49]. China remains the dominant producer and exporter, supplying over 80% of the global market [50]. However, China's REE exports declined by 4.4% year-on-year in the first five months of 2023 [51]. Since 2014, REEs' prices have fluctuated, with La-oxide, Nd-oxide, and Y-oxide prices rising significantly by 2018 [52].

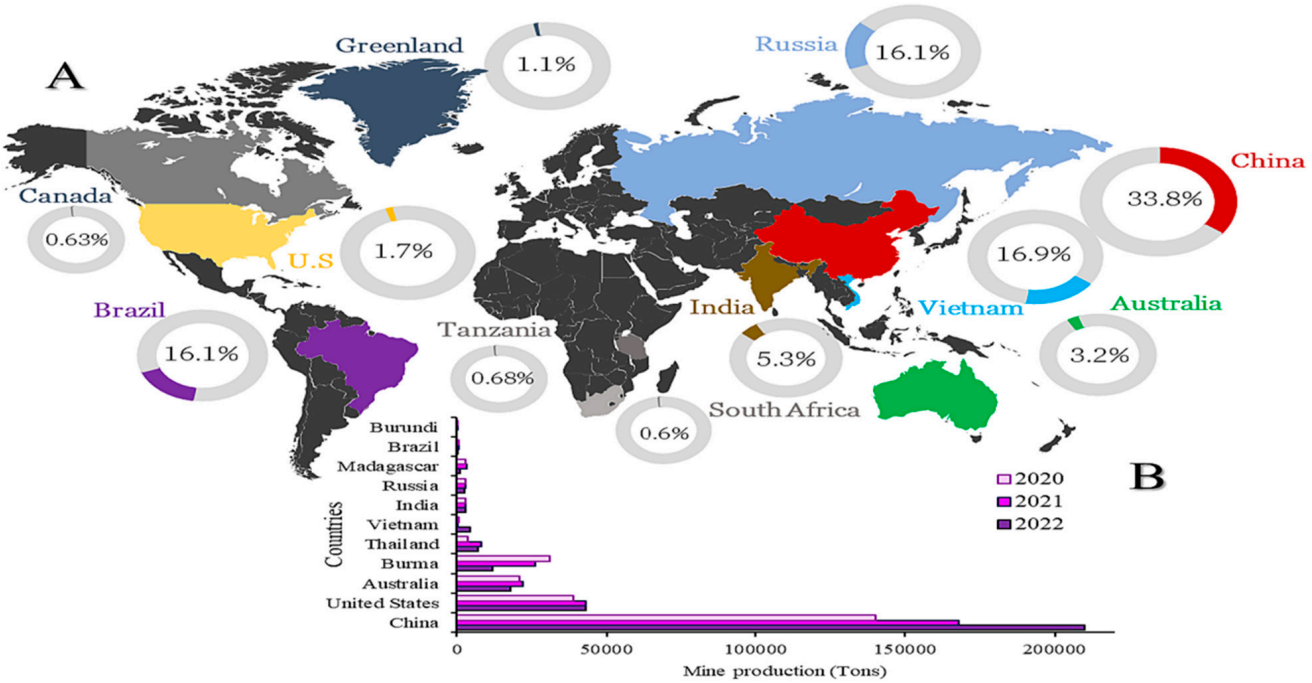


Figure 2. Global REEs reserves (**A**) and annual production (**B**). Data were reproduced from U.S. Geological Survey, 2023. Reproduced with permission from Ref. [46].

The environmental effects of REEs are significant. The mining and extraction processes pose risks to ecosystems and human health, causing geological disruptions, soil erosion, and hydrogeological changes. These activities release harmful pollutants, contaminating air, water, and soil, and rare earth ores often contain radioactive thorium and uranium, which pose severe health risks. Additionally, REE production leading to further pollution and environmental strain. Managing and disposing of hazardous chemical byproducts from production is challenging. Consequently, recycling rare earth elements from resources, such as end-of-life materials, is crucial to mitigating environmental impacts and ensuring a reliable domestic supply [46].

As widely used materials, REEs exhibit significantly high concentrations in the waste from electrical and electronic equipment [40]. The development of recycling processes is crucial due to the unique challenges presented by each waste stream, aiming to achieve economically viable extraction. The cost of processing REEs is attributed to the low concentration of rare earth ore deposits, the chemical similarities among elements, and the need for high-purity single-element products. The economically inefficient mining of specific elements makes it unfeasible to selectively target a single element [53]. The prices of the mined elements might vary, with elements such as cerium often being provided at prices

below cost, while compensating by setting higher prices for other elements, maintaining an overall positive return for REE producers. A surge in demand for specific REEs may trigger an overproduction of less-demanded elements [5]. The classification of "criticality" is determined by the importance of rare earth elements (REEs) for specific applications, such as renewable energy, the absence of comparable and reliable substitutes, and the monopolization of supply sources. According to the US Department of Energy, the category of critical REEs encompasses five elements: neodymium, europium, terbium, dysprosium, and yttrium. This designation of "critical" is often used as a benchmark in project feasibility studies, as ore bodies containing higher proportions of critical REEs are perceived to be less vulnerable to market fluctuations [5].

Moreover, the expansion of the REE industry gives rise to concerns regarding industrial health issues. Exposure to REEs is a possibility in multiple stages of the industry, such as mining, processing, transportation, disposal, and decomposition [54]. Exposure of humans to REEs takes place through inhalation, ingestion, and skin contact, with notable exposure occurring in recycling facilities where workers encounter substantial amounts of REEs. Prolonged exposure to REEs has been linked to conditions such as pulmonary fibrosis and pneumoconiosis, and studies have reported in vivo/in vitro toxicities associated with REEs [55]. Emissions of harmful substances during both REE mining and processing, coupled with the analogous processes involved in REE recycling, have the potential to lead to health and environmental issues [37]. The complexity of these issues transcends disciplines.

3. Reuse and Recycling of REEs

The widely adopted waste management hierarchy follows a global framework, starting with a technique that involves reducing waste at its source. This primarily involves the reduction of excess waste, whether it be in the form of material packaging or optimizing material use. The second step in the hierarchy emphasizes reuse, encouraging the repeated use of materials after the end of their lifecycle. Following that, the third option involves recycling or composting materials. Recycling includes finding new purposes for materials that cannot be reused, often transforming their form. The fourth step is recovery, which involves processing waste in some way to generate a valuable result, such as recovering energy and metals from the waste. Disposal is the last step, where waste without current value is discarded in a secure manner [56].

3.1. Reuse of REEs

REEs play a crucial role in advanced electronics and diagnostic and scanning equipment in various fields, due to their highly magnetic, conductive, phosphorescent, catalytic, electrochemical, and luminescent properties [57,58]. Consequently, REEs have become indispensable in modern sophisticated lifestyles. They are essential in metal refining, alloy manufacturing, petrochemical processing, locomotive and automobile catalysis, superconductors, coloring pigments for glass and ceramics, fuel cells, military equipment, permanent magnets, and laser light manufacturing. Additionally, REEs are widely used in household appliances such as rechargeable batteries, mobile phone speakers, microphones, fiber optic cables, compact fluorescent lamps, LED lights, and hard disk drives [12].

A technology designed for a circular economy should focus on minimizing materials and energy wastage, while encouraging the reuse and recycling of resources. In the context of recycling, which involves the reuse and recovery of REEs, decisions must be made regarding whether to directly reuse or recover the contents. Creating scalable and sustainable effects within circular economy business models (CEBMs) presents numerous challenges. CEBMs focus on closed-loop manufacturing, material reuse, prolonging product lifespan, and devising innovative recycling techniques to extract the inherent value of materials. Closed-loop cycles come in two forms: upcycling and downcycling [59]. In up-cycling, materials at the end of their lifespan are reused to produce a higher-value outcome, and in down-cycling, these materials are reused to produce a product of lower

value [59] Recycling models are built upon closed-loop cycles, offering opportunities to optimize inputs, foster successful elements, and grasp the dynamic impacts of evolving CEBMs. Upcycling contributes significantly from a sustainability standpoint within the sociotechnical system [59].

In a resource-efficient economy, recycling and reusing metals are imperative. Products reaching the end of their life cycle that contain REEs should ideally undergo preprocessing to yield recycled materials rich in REEs, which can then be later processed to generate rare earth compounds, facilitating their reuse in subsequent production cycles [3]. Preprocessing involves disassembly, physical separation, and size reduction. When recovering metals and other REEs from electronic waste, selecting suitable technology poses a challenge due to variations in element type, metal content, and composition. Enhancing the technical efficiency of the recovery process with a material-centered approach while minimizing pollution is crucial. However, current preprocessing technologies suffer from drawbacks such as the loss of critical elements, like shredding dust. Therefore, optimizing the pre-treatment stage is imperative to prevent REEs loss during recovery. Integrating pre-treatment processes with chemical and other physical methods is also necessary for energy recovery and the removal of hazardous components [40]. The most economical recycling method is direct reuse, characterized by low energy input, no consumption of chemicals, and the absence of waste generation [3]. While recycling and reusing end-of-life REE products are feasible, the processes involved are complex and consume a lot of energy [60]. Directly reusing end-of-life magnets is frequently unfeasible due to preprocessing needs, including the removal of coating and glue residues, the risk of magnetic properties loss, and the design-specific shapes tailored for individual products [61].

3.2. Recycling of REES

Since the 1970s, recycling technologies for REEs have gained significant attention, driven by their economic and environmental advantages. Recycling stands out by offering several benefits compared to primary production, particularly by circumventing the complexities associated with separating radioactive elements Moreover, recycling REEs from waste materials plays a role in diminishing their environmental impact [62]. There are various approaches to recycling and recovering REEs [35], underscoring the need for process developments to address the distinct challenges posed by each waste stream. Research groups globally have been actively working on the development of recycling processes for secondary streams, as extensively reviewed in the literature [17,63]. These approaches are presented in Table 2.

Table 2. Advantages, drawbacks, risks and costs of recycling technologies of REE recovery.

Recycling Methods	Advantages	Drawbacks	Hazard/Risk	Cost
Hydrometallurgical process (leaching, solvent extraction, precipitation,)	High-purity levels for individual REEs and direct scalability [64].	Involve numerous steps and the use of various chemicals before obtaining a newly recycled magnet [15].	Substantial amounts of wastewater that require appropriate treatment to prevent environmental pollution [65]	Costs related to the high energy, reagent, and capital requirements [62].
Pyrometallurgical process (roasting, calcination)	Absence of liquid waste byproducts, potential to obtain (REMs) and a high level of reagent regeneration [66].	Require large energy inputs to bring enough heat to the material [67].	The utilization of high temperatures can result in the emission of harmful gases and dioxins, as well as the generation of solid wastes [37].	Necessitate substantial investments when compared to hydrometallurgy [68].
Bioleaching	Lower environmental footprint compared to other REE recycling methods [18].	Slow kinetics and lower recoveries [69].	Produce undesirable acid mine drainage [70].	Cost effective [71]
Step by step	Size-selective crystallization, only requires one step of operation, and can obtain a single REE [72]	The process features extended operational cycles, significant REEs losses, subpar separation outcomes, and remains relatively underutilized [73].	Environmentally friendly [72].	Low cost [72].
Solvent extraction	Large processing capacity, excellent selectivity, and elevated product purity [72].	Requiring multiple extractions to attain optimal purity [72].	Use of hazardous (i.e., organophosphorus) chemicals [74]	Extractants choice is impacted by cost [75]
Supercritical fluids	With low viscosity, high diffusivity, and solvation properties, they prove to be efficient solvents for metal extraction [76].	High temperature and pressure reactors [77].	Supercritical CO_2 is non-toxic, non-flammable, and inert, making it environmentally acceptable [78].	Supercritical CO_2 is inexpensive [78].
Ionic fluids	Comprising molten salts, they offer an alternative to organic reagents in liquid–liquid extraction due to their exceptional extractability [79].	Partial dissolution in the aqueous phase [80].	Toxicity in the preparation and process [79].	Resin material is poor in regeneration and usability, resulting in high process costs [72].
Membrane separation	Effective, straightforward, energy-efficient, and capable of minimizing organic solvent waste [81].	The limited availability of appropriate membrane materials restricts the utilization of the technology for purifying rare earth ions [72].	It represents a comparatively environmentally-friendly approach to REEs separation [81].	High cost and membrane replacement [14].
Adsorption	Has a broad concentration range [82].	Pre-concentration of adsorbents for industrial-scale use remains a challenge for bio-adsorbents [72]. Silicon-based materials exhibit inadequate stability in acidic environments [83]. Capacity and selectivity of magnetic adsorption for rare earth ions in aqueous solutions are suboptimal [72].	Causing secondary wastes [84].	Additional operation costs [84].
Electrochemical process	Environmental conservation, minimal energy usage, ease of operation, and sustained functionality [72].	If the metal must remain in a liquid state, the electrolyte must possess a melting point higher than that of the metal, which can limit the options for selecting the electrolytic cell and electrolyte [72].	Formation of solid Fe-based solid waste [85].	Cost effective [86].

To address REE supply challenges, three strategies can be proposed: diversifying supply sources, developing substitute materials, and promoting the reuse, recovery, and efficient utilization of materials in manufacturing processes. Recycling REEs from waste materials minimizes environmental impacts. Different REE recycling methods are available, including commonly used techniques such as hydrometallurgy, pyrometallurgy, and selective electrochemical recovery. Aside from these conventional methods, alternative options like biometallurgy are gaining traction in the industry [46]. Currently, the primary methods employed for extracting REEs are pyrometallurgy and hydrometallurgy [87].

3.2.1. Pyrometallurgical Methods

Pyrometallurgy provides an alternative to mitigate the high water consumption and production of dangerous waste, even though it requires a lot of energy and is still undergoing research [88]. Pyrometallurgy stands out as a recognized thermal treatment for reclaiming metals from electronic waste [40]. Müller and Friedrich [89] explored the efficiency of CaO–SiO_2 and CaO–CaF_2 fluxing systems in extracting REEs from NiMHBs. Their research revealed that the CaO–SiO_2 system exhibited limited separation between metal and slag due to the high viscosity of the slag, resulting in silicon enrichment in the metal phase. In a separate study, Hsu et al. [90] reported a 70% recovery rate of REEs from electronic wastes, such as PCBs, through a pyrometallurgical process in 2019. This method, known for its high temperatures, is often considered more environmentally sustainable due to reduced water usage and minimized hazardous waste production. However, it does entail higher energy consumption. Despite these advantages, certain pyrometallurgical processes still necessitate chemical inputs and result in waste generation. While pyrometallurgical recycling enables scalability and involves fewer chemicals (flame retardants, furans, and phenolics) [16]), it does release harmful gases like dioxins, carbon dioxide, and volatile metals such as mercury [91]. Cui and Zhang demonstrated that polychlorinated biphenyls (PCBs) can generate a mixed oxide, primarily containing lead (Pb) and zinc (Zn), as slag through a top-blown reactor. They also observed the formation of a copper–nickel–silicon alloy, where high pressure facilitated the separation of bismuth (Bi), antimony (Sb), lead (Pb), and other heavy metals. However, this process faces certain limitations concerning the pillars of the UN's sustainability goals. One issue pertains to environmental concerns linked to the release of toxic substances, dioxin formation, and advanced smelting practices, which may result in slag production and additional industrial systems. Another challenge involves the necessity for a more sophisticated emission control system, which incurs higher costs [92]. For instance, some liquid-phase techniques like electroslag refining produce significant solid waste, while certain gas-phase methods require substantial amounts of chlorine gas. Pyrometallurgical extraction encompasses various techniques, including electrochemical processing, gas-phase extraction, gas–solid and solid–liquid processes, glass slag processing, roasting, molten slag extraction, liquid-metal extraction, molten salt extraction, and smelting [63]. Among these, smelting is commonly employed for the extraction of rare earth element oxides, which are subsequently dissolved in a slag phase [93]. Smelting boasts rapid kinetics and facilitates easy separation of residues [94].

3.2.2. Hydrometallurgical Methods

Hydrometallurgy is a chemical approach for extracting metals from electronic waste, demonstrating high efficiency in metal recovery through precipitation and ionic liquid techniques. However, metal recovery via precipitation often results in the formation of secondary sludge [40]. This typically leads to the generation of substantial liquid waste volumes and significant environmental impact. Nonetheless, novel separation methodologies rooted in green chemistry, closed-loop systems, and coordination chemistry are currently in development to mitigate the environmental impact of NdFeB magnet recycling via hydrometallurgy [95]. Various hydrometallurgical methods have been employed, including liquid metal extraction, molten slag extraction, liquid media extraction, and supercritical fluid extraction [96]. Common procedures entail selective leaching of WEEE

(Waste Electrical and Electronic Equipment) using mineral acids, followed by adsorption, ion exchange, precipitation, solvent extraction, or supported liquid membrane separation [63,97]. A hydrometallurgical process involves a series of chemical operations aimed at: (i) leaching one or multiple targeted elements from the ore or waste (typically serves as the initial step following mechanical or thermal pre-treatment [98]), (ii) purifying the solution by removing impurities, and (iii) extracting the targeted element(s) from this solution [99].

In the case of hydrometallurgy, rare earth elements (REEs) like gadolinium, yttrium, and lanthanum are extracted from electronic waste using a chloride solution. Previous studies have highlighted certain drawbacks of recovering these elements from electronic waste, including sludge generation, heavy metal pollution, and toxicity [100,101]. However, research by Tuncuk et al. demonstrated that hydrometallurgical processes could effectively recover metals from printed circuit boards, though with limitations as it primarily recovers only lithium and cobalt metals, omitting others [102].

Irrespective of the selected method, the retrieval of REEs involves leaching and separation processes, followed by purification [103]. The leaching process of metals from electronic waste can be influenced by various factors such as redox potential, leaching kinetics, lixiviant type, particle size, pH, temperature, and agitation [104]. Furthermore, several studies have suggested that metal recovery through electro-winning and adsorption methods could address the issue of sludge production [105]. REE production frequently entails environmental concerns. Processes such as mining, leaching, pre-concentration, and the multiple stages of separation necessary to attain the levels of purity demanded in certain applications, occasionally reaching up to 99.999%, commonly result in substantial quantities of secondary waste [106]. For instance, a solid-state chlorination process, succeeded by leaching with an organic solvent, was employed to selectively recover Y (95.7%) and Eu (92.2%) from fluorescent lamp wastes [107]. Sulfuric and hydrochloric acids were used to leach out over 97% of Eu and Y from computer monitor waste [108]. Other works have used modified ionic liquids to selectively leach REEs from fluorescent powder, Nd-Fe-B magnets, and NiMH battery waste [109]. The effectiveness of acid leaching in recovering metals from various matrices has been well documented [110]. Another study using 5 M HCl leached out Dy (76%), Nd (78%), and Pr (83%) from waste permanent magnets [111]. Sulfuric and hydrochloric acids achieved over 97% Eu and Y recovery from computer monitor waste [108]. Additionally, modified ionic liquids have been used to selectively leach REEs from fluorescent powder, Nd-Fe-B magnets, and NiMH battery waste [109].

In a study by Korkmaz and colleagues [90], a total recovery of up to 86% for rare earth elements (REEs) from a mixed hydrated acid (MHA) leachate was achieved, with minimal co-precipitation of other elements observed. The REEs were precipitated as mixed hydrated sulfates by the addition of ethanol and 2-propanol as antisolvents, as reported by Korkmaz. However, despite the high yield of REE recovery, a significant volume of alcohol was required for effective precipitation [112]. A novel phosphate-based extraction–precipitation strategy was developed for recovering rare earths (La, Ce, Nd, and Pr) from waste nickel–metal hydride (NiMH) batteries, eliminating the need for saponification and organic solvents. Dibenzyl phosphate (DBP), diphenyl phosphate (DPP), and triphenyl phosphate (TPP) were tested as extraction-precipitants. DBP showed the highest precipitation efficiencies for RE3+ at 97.84%, 100%, 100%, and 99.77%, respectively, while maintaining low precipitation efficiencies for Mn^{2+}, Co^{2+}, and Ni^{2+} (less than 1.75%). The larger particle sizes of DBP-RE precipitates improved solid–liquid separation efficiency. Using 3 mol/L hydrochloric acid, the stripping efficiency of DBP-RE reached 98.60%, with a recovered REE purity of 99.85% [113].

A sustainable method was developed for efficiently separating and recovering REEs and iron from NdFeB slurry. REEs were recovered via one-step selective precipitation in phosphoric acid, while iron was recovered using oxalic acid. Using 4 mol/L phosphoric acid at 80 °C with a liquid-to-solid ratio of 30:1 for 90 min, the leaching efficiencies were 98.76% for Fe and 1.09% for REEs. The remaining REEs were retained as $REEPO_4 \cdot nH_2O$ in the residue. Mixed rare earth oxide (from roasted rare earth oxalate) and $FeC_2O4 \cdot {}_2H2O$

with purities of 99.49% and 97.17%, respectively, were obtained [114]. Additionally, a method for separating metals from waste mobile phones using aqua regia leaching and pH-based precipitation was described, with iron precipitated at pH 3.1–4.2, copper at pH 5.7–7.7, and rare metals at pH 8.3–10.5 [115].

REE extraction and recovery via hydrometallurgical methods involve three sequential steps. A flowchart for REE separation is provided in Figure 3.

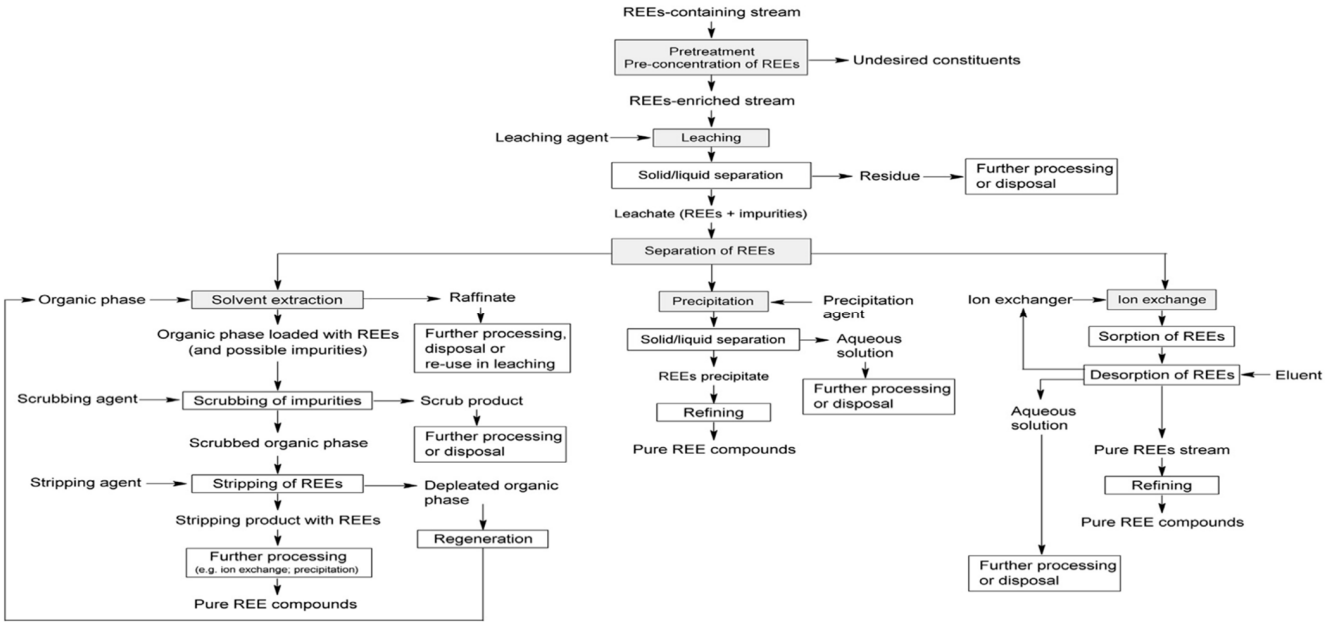


Figure 3. A simplified flowchart outlining pre-treatment, leaching, solvent extraction, ion exchange, and precipitation methods for the separation of REEs [106].

- Assessment, evaluation, and initial processing of the material;
- Extraction and separation of metal ions through leaching;
- Purification of the resultant product to achieve the desired form, either as pure metal or pure metal compounds.

The execution of these three stages presupposes thorough collection and sorting of the products beforehand. This preliminary process is essential for the efficient treatment and recovery of the contained fractions [106].

3.2.3. Approaches for Separation of REEs

The demand for substantial quantities of pure REEs is compounded by the inherent challenge in separating them, as they possess closely analogous physicochemical properties. Furthermore, existing purification and separation methods for REEs contribute to environmental impacts, including the generation of undesired waste, the discharge of used organic solvents, and excessive water consumption, thereby constraining the widespread extraction of REEs [116].

Various purification processes can be employed to recover precious metals present in the leachates. Solvent extraction, chemical precipitation, ion exchange, adsorption, electrochemical deposition, and more innovative eutectic freeze crystallization are among the most common separation methods [91,98]. Although precipitation and ionic liquid methods are highly efficient for metal recovery, the use of precipitation often results in the generation of secondary sludge, which can lead to pollution [40]. Consequently, these methods face challenges hindering their widespread industrial application [91]. Generally, ion exchange and solvent extraction, although more intricate and costly, are employed for obtaining pure individual REE products, whereas selective precipitation, which is more cost-effective, is preferred for generating mixed REE products [117]. However, studies have

demonstrated that metal recovery through electro-winning and adsorption could mitigate the issue of sludge production [40].

Bogart and colleagues [118] introduced an organic compound capable of binding to REE cations, facilitating the formation of distinct compounds for LREEs and HREEs. This resulted in the LREEs forming dimeric aggregations, while the HREE compounds did not aggregate. Such disparity in solubility enabled effective separation of LREEs and HREEs through filtration [118]. Further refinement of this separation process led to the creation of specific pairs of REEs, such as Nd-Dy and Eu-Y, essential in manufacturing permanent REE magnets and fluorescent bulbs. If scalable, this novel approach to REE recycling could represent a significant stride towards enhancing the recyclability of various REE-bearing end products [18]. It is essential to recognize that recycling is just one of several potential responses to perceived REE supply risks. Peelman et al. [119] have underscored the potential for extracting REEs from mine tailings, exemplified by the Kiruna iron ore mine in Sweden. Recovering REEs from acid mine drainage could yield economic, environmental, and strategic advantages. Similarly, exploration of alternative recycling methods should be encouraged; for instance, bioleaching offers a markedly lower environmental impact compared to other REE recycling techniques [18].

Step by Step Method

Differences in the solubility of REEs are utilized by introducing different chemical reactants to aid in separation through crystallization and precipitation. These techniques leverage the ability of rare earth ions to form complexes with various ligands, thereby expediting the separation process. However, because the size differences among rare earth elements are minimal, conventional stepwise methods necessitate numerous dissolution-precipitation cycles to isolate a single element. This renders the process impractical for large-scale industrial production due to the extended production cycles and associated high costs.

While the separation efficacy of rare earths has seen enhancement, the stepwise method encounters challenges due to minimal solubility differences among adjacent rare earths. This results in prolonged operation cycles, significant rare earth losses, and suboptimal separation outcomes, hence limiting its widespread adoption. With adjacent rare earth ions exhibiting a mere 0.03 A radius difference, relying on size disparity for separation proves exceedingly arduous [72]. In a study by Zhao Xiang et al. [120], they utilized cost-effective camphoric acid and lanthanides to synthesize organic rare earth metal framework materials, highlighting the significant potential of these materials in separating individual REEs. These organic rare earth metal frameworks (MOFs) present diverse structures and properties, marking a groundbreaking advancement in rare earth element separation [120]. Since MOFs consist of metal complex polymers, it is proposed that even a slight difference in Kf for a molecular Ln^{3+} complex could be magnified during MOF crystallization, resulting in pronounced differences in the ability of each Ln^{3+} to form a specific MOF [120]. Their approach involves the selective crystallization of Ln^{3+} ions into MOFs, employing a blend of D-camphoric and formic ligands to achieve the targeted crystallization of specific lanthanide ions into MOFs, thereby establishing distinct categories within the MOF structure [121]. Throughout the formation of MOF crystal frameworks, variations in the size of rare earth ions facilitate increased repulsive forces between interconnected ligands, leading to highly selective size crystallization and efficient single rare earth element separation [72].

The size-selective crystallization method achieves quantitative separation by controlling reaction kinetics, making it efficient and cost-effective for industrial applications [72]. In order to reduce radioactive impurities in rare earths, Wang et al. refined the differentiation between trivalent thorium and tetravalent lanthanides [122]. This work demonstrates, for the first time, the substantial potential of these materials in effectively isolating individual REEs as shown in Figure 4a–f) [120]. Yang et al. introduced a Valence Selective Crystallization (VSC) approach, utilizing Ce as the target ion (Figure 4g) [123]. The technique achieved an impressive purification efficiency of 99.97% for Ce in the mixed solution,

and scale-up experiments further demonstrated effective selective separation of target ions, suggesting potential for industrial applications [123]. This approach not only emphasizes environmental friendliness and energy efficiency but also achieves precise isolation of mixed Ce. Considering that the majority of REEs exist in the trivalent state, except for Ce, which can be tetravalent, this method proves particularly suitable for capturing tetravalent ions. Yin et al. accomplished the separation of individual REEs by controlling kinetics of binary REE models. Their findings demonstrate that borate crystallization enables specific identification of REES (Figure 4f) [124].

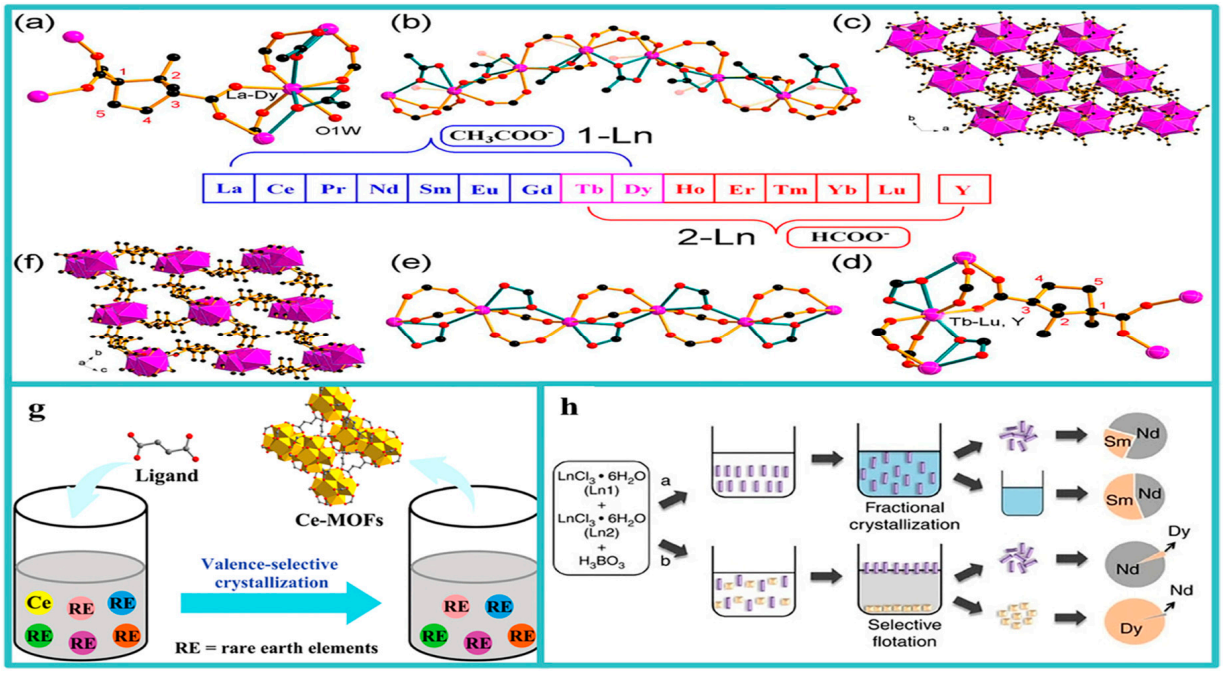


Figure 4. Graphical representation of (**a**–**f**) selective crystallization of lanthanides [120], (**g**) separation of Ce^{4+} in REE solution using VSC [123], and (**h**) selective crystallization and separation of lanthanides. Reproduced with permission from Ref. [72]. In this investigation, distinct ions interacted with the identical ligand to form varying crystals, which can then be separated through flotation. This marks another significant progression in the separation of REEs through size-dependent crystallization techniques [72].

Operating in a single step, this method accomplishes the full separation of individual REEs by regulating reaction kinetics. Subsequently, distinct crystalline products of various REEs are isolated through flotation, introducing a novel approach to rare earth separation based on ion size. However, further research is necessary to comprehensively grasp the mechanism of size-selective crystallization of MOFs [72].

Ionic Liquids

The stepwise method, while suffering from low purity, prompted the development of the ion exchange method as a remedy for these shortcomings [72]. Several techniques have been suggested for the recycling of lamp phosphors. An innovative method involves the use of ionic liquid (IL), which can be regenerated and reused [125]. The significant role of ionic liquids in REEs was emphasized by Kaim et al., highlighting the stable reusability of bifunctional ionic liquids even after multiple experiments, with negligible loss of extractants [126]. ILs have seen a growing application in solvent extractions to improve the separation capabilities of established extractants as eco-friendly alternatives to conventional organic solvents. This trend is owed to their advantageous attributes, including low vapor pressure, minimal flammability, the capacity to dissolve both organic and inorganic compounds, high thermal stability, elevated conductivity, and broad electrochemical win-

dows. Furthermore, ILs can influence separation processes through solvent interactions and ion-exchange mechanisms, thereby enabling the customization of specific extractant properties. Consequently, there has been a heightened interest in extraction techniques employing functionalized ILs, also referred to as task-specific ionic liquids (TSILs) [80]. Ionic solvents can be defined as salts with melting points below 100 °C possessing modular physical and chemical characteristics [23]. They have attracted research interest due to their broad applications, serving as an environmentally friendly alternative to organic solvents in diverse synthetic reactions [126]. Recent research suggests that the toxicity of ILs is closely tied to the structure of the IL, particularly the cationic component [127].

Figure 5a illustrates the use of ion exchange resin for recovering Dy and Nd from waste magnets, resulting in purities of 98.5% and 91.6%, respectively [128].

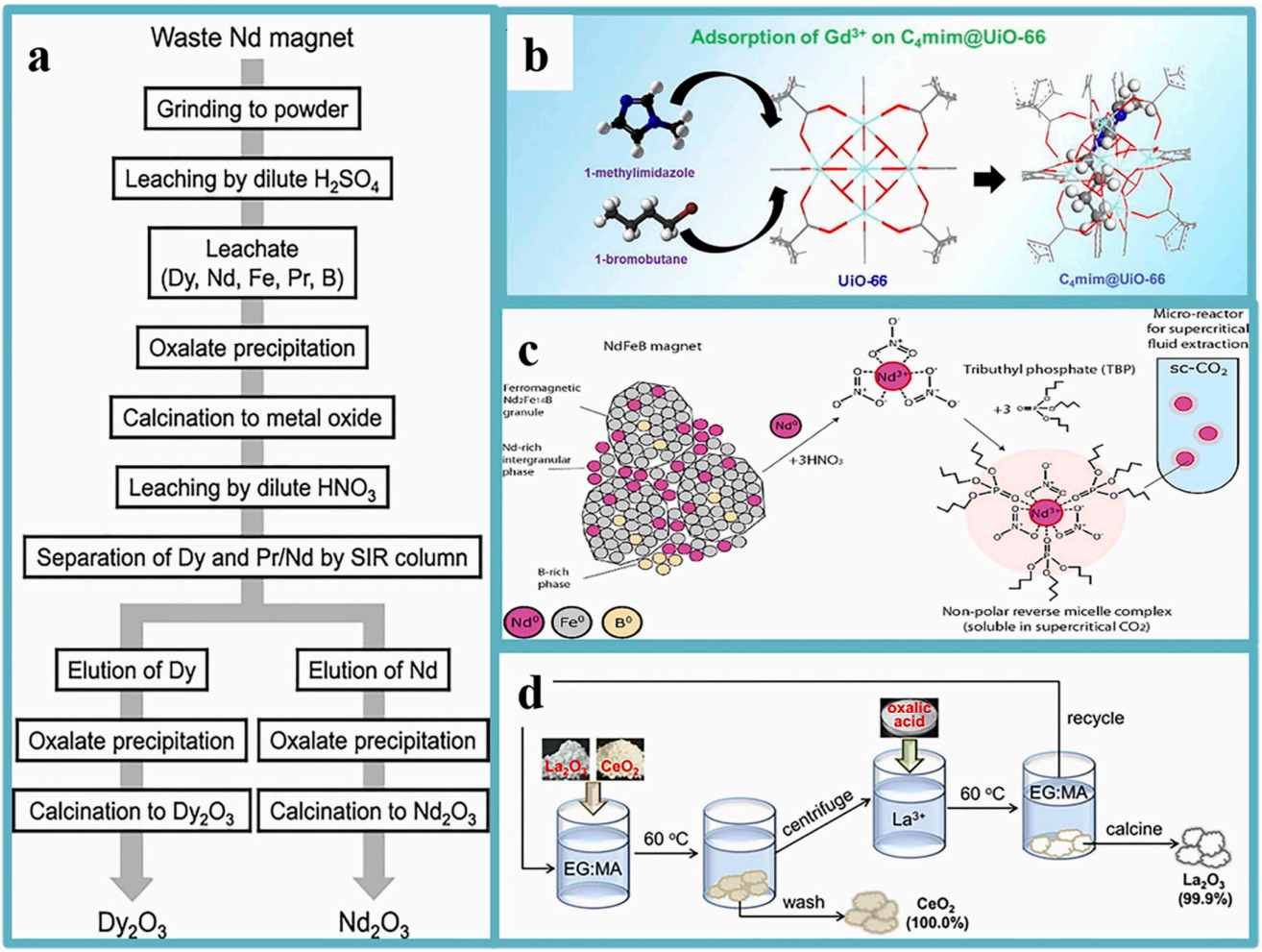


Figure 5. (**a**) Recovery process for Dy and Nd from waste Nd magnets is depicted in a comprehensive flowchart [128]. (**b**) Ionic liquid-coated UiO-66 demonstrates the capture of Gd^{3+} in solution [129]. (**c**) Model illustrating using supercritical carbon dioxide for the extraction of REEs [130]. (**d**) Separation process visually presented [131]. Reproduced with permission from Ref. [72].

Ahmed et al. utilized a technique to encapsulate the ionic liquid within a stable hollow cage structure [129]. The inclusion of the ionic liquid in this process enhances the adsorption capability of the adsorbent, leading to a selectivity of more than 82%, as shown in Figure 5b. Zhang et al. utilized tributyl phosphate–nitric acid with methanol to improve the interaction between solute and the solvent, as depicted in Figure 5c. Their results provide evidence of the potential for separating and recovering REES using supercritical extractants [130]. Chen et al. developed eco-friendly Brønsted acidic deep eutectic solvents (DESs) using

carboxylic acids and polyols. These DESs effectively utilize the differing solubility of metal oxides, demonstrating purification rates of 99.9%, as indicated in Figure 5d [72,131].

The ion exchange method offers the capability to separate various elements in a single operation, boasting advantages such as minimal pollution and high product purity. However, it suffers from a prolonged production cycle due to the poor regenerative capacity and limited continuous processing capability of resin materials, leading to elevated process costs. Although not currently the primary technique for rare earth separation [132], there is a necessity for additional exploration of ion exchange technology, primarily for producing high-purity rare earth elements. Integrating ion exchange with other separation methods can be beneficial, such as substituting extractants with ionic liquids in solvent extraction techniques or using polymeric materials coated with ionic liquids to enhance ion exchange processes [128,133]. Yamada et al. developed a chromatographic medium to minimize extractant loss. This medium successfully recovered Dy and Nd from discarded neodymium magnets, with purities of 98.5% and 91.6%, respectively [128]. Solvent-impregnated resins (SIRs) can be lost within the carrier matrix in industrial settings. To address this issue, Yamada et al. developed a method where they impregnated a support material to prevent extractant loss. This chromatographic medium was utilized to extract Dy and Nd, resulting in purities of 98.5% and 91.6%, respectively [128].

The complexity of ionic liquids (ILs) corresponds to the unpredictability of their mechanisms. The utilization of ILs in REE extraction has become increasingly pressing due to the growing demand for environmentally friendly practices in the REE separation process. Traditional REE extraction methods typically involve significant consumption of volatile organic solvents (VOCs), leading to both short- and long-term adverse effects. Moreover, the expenses associated with managing VOCs have escalated due to stricter regulations. In ion exchange methodologies, in order to improve the selective separation efficacy of REEs, selecting suitable ion exchangers and optimizing adsorption and leaching conditions is required. Variables such as cross-linking as well as the capacity of exchange sites greatly influence the exchange process [134].

The process of cation exchange involves the simultaneous migration of the cationic component of ILs to the aqueous phase, which poses limitations on its practical application in large-scale processes. Rout et al. [135] confirmed this hypothesis through their research, comparing the extraction processes involving ILs with longer alkyl chains, such as [C10mim][Tf2N], [P66614][Tf2N], or [N1888][Tf2N], with those having shorter alkyl chains like [C4mim][Tf2N] or [N1444][Tf2N]. As anticipated, the cation exchange was inhibited in ILs with longer alkyl chains, leading to a reduction in extraction efficiency as the hydrophobic cation encountered greater difficulty in migrating to the aqueous phase. Consequently, fewer REE complexes were extracted, highlighting the need for further research to elucidate how the elongation of the alkyl chain diminishes extraction efficiency [136]. The recycling processes vary, often based on the hydrophobicity/hydrophilicity of an IL. Nonetheless, a primary objective of these processes is to reuse the ILs in subsequent cycles. One common method for the reuse of ILs, particularly applicable to hydrophilic thermally stable ILs, is thermal treatment. Other techniques include centrifugation, adsorption, reverse osmosis, and electrodialysis [137]. Typically, an IL-based extraction process begins with an aqueous solution of REE ions interacting with an IL layer containing the extractant. The REE ions are chelated by the extractant and IL cations/anions, migrating from the aqueous phase to the IL phase. Following layer separation, the IL phase can undergo a stripping process, transferring the REEs to a solution with higher affinity, often an acidic aqueous solution containing nitric acid or hydrochloric acid. The remaining IL phase can then be reused for subsequent extractions [80]. However, the chemistry of rare earths-based ionic liquids remains largely unexplored [138].

ILs are regarded as a more environmentally friendly alternative to traditional volatile organic solvents (VOCs) due to their lower vapor pressures [137]. Despite their designation as ‘green’ solvents, their solubility in water can potentially lead to aquatic contamination.

Additionally, studies have shown that the overall toxicity of ILs depends on both the cation, particularly the alkyl chain length, and the anion components [23].

Ion exchange resins, which consist of strongly acidic cation exchangers and strong basic anion exchange resins, are utilized as the exchange medium to selectively separate different rare earth elements. To improve the selectivity of the separation process in ion exchange techniques, the selection of suitable ion exchangers and the optimization of adsorption and leaching conditions are crucial. Variables such as the type of exchange group, extent of cross-linking, and size of exchange capacity play a significant role in influencing the efficiency of the exchange process [139]. Chelation between rare earth ions and ionic liquid (IL) species enables the transfer of chelated rare earth elements from the aqueous phase to the IL phase. However, there are cases where both the IL cations and anions can migrate into the water phase, resulting in IL loss and impeding efficient recycling. To address this, combining ILs with polymer materials proves effective in reducing IL loss. Polymer materials are loaded with ionic liquid extractants through cross-linking reactions, forming a film that contains the IL. This film, when combined with techniques such as ion exchange resin, chromatography, and ICP-MS, facilitates the separation of solutions with low REE content [72].

Ideally, ILs should be used in a way that allows for regeneration or recycling, thereby significantly lowering expenses. Wang et al. suggest IL-based rare earth element (REE) extraction processes should be designed with this goal in mind [136].

Solvent Extraction

In comparison to the ion exchange technique, solvent extraction allows for the swift and continuous extraction of REEs [72]. Solvent extraction has gained prominence due to its substantial processing capacity, rapid mass transfer rate, and excellent separation selectivity, gradually supplanting conventional approaches. Presently, solvent extraction stands as the primary and crucial technology for the separation of REEs [140].

Liquid–liquid extraction stands as the prevalent method for separating mixed REEs. In this procedure, REEs are selectively extracted from the leach solution by an organic solvent, capitalizing on their unique chemical attributes like solubility and affinity for the solvent [97].

Solvent extraction involves the use of a solvent, also known as the organic phase, composed of an extractant dissolved in a diluent. While pure extractant is occasionally utilized, it is more common to employ a diluent, as many extractants exhibit high viscosity in their undiluted state. Additionally, a modifier may be introduced into the organic phase to prevent the formation of a third phase [141]. The extractant plays a crucial role in solvent extraction, leading to the synthesis and documentation of numerous extractants for both analytical and commercial purposes [142]. In the case of HREEs, their extraction capability surpasses that of LREEs. However, the ease of extraction may correspond to a more challenging stripping process. Consequently, when selecting extractants for HREEs, achieving high stripping efficiency while appropriately reducing extraction capability is considered a more resource-efficient approach [140]. The principle behind extraction lies in the distribution of REEs between the two immiscible phases. Pure REEs are subsequently recovered through stripping or back extraction utilizing an acid, base, or water solution [107].

Oxalic acid is commonly utilized as a precipitating agent to separate REEs from the leach solution, owing to its strong binding affinity to REEs across a broad pH spectrum [143]. Organophosphorus extractants such as di-(2-ethylhexyl) phosphoric acid (D2EHPA) and tributyl phosphate (TBP) are extensively employed in REE extraction due to their high selectivity for heavy rare earth elements (HREEs) [144]. Ionic liquid extractants like Aliquat 336 exhibit proficiency due to their elevated selectivity [145]. Cation exchange agents are extractants with acidic functional groups which selectively bond to positively charged rare earth elements (REEs) [139]. Saratale et al. [146] found that Versatic Acid 10, an organic carboxylic acid extractant, is effective in separating mixed REEs. Bie et al. investigated the use of three novel oxyacetic acid extractants (BPAOA, BEAOA, and DBOA) [147] for the separation of

thorium and rare earths, addressing the issue of secondary entrainment in the hydrochloric acid system. Riaño and colleagues employed the ionic liquid trihexyl(tetradecyl) in the successful separation and recovery of Nd_2O_3 and Dy_2O_3 from waste NdFeB permanent magnets, achieving purities of 99.6% and 99.8%, respectively [148].

The utilization of supercritical extraction technology has been explored for REEs separation [72]. Supercritical fluids exhibit advantageous extraction characteristics, including low viscosity, high diffusivity, and efficient solvation capabilities, rendering them highly effective solvents for metal extraction. Several extraction agents have been studied for supercritical rare earth extraction [149]. Zhang et al. aimed to improve the interaction between the solute and solvent. Their findings demonstrated the potential of supercritical extractants for the separation and recovery of REEs [130].

Among these, supercritical carbon dioxide (sc-CO_2) stands out as the most commonly employed solvent due to its moderate critical temperature (Tc = 31.1 °C) and pressure (7.37 MPa). Furthermore, sc-CO_2, offers additional benefits such as inertness, accessibility, affordability, and ease of venting or convenient recyclability through phase separation from products post-extraction and depressurization [76]. Rare earth elements (REEs) are extracted from electronic waste through a combination of supercritical water and acid leaching (HCl). However, one drawback of this integration is the necessity for high temperatures (approximately 420–440 °C), which poses a challenge. To address this issue, there is a growing focus on developing greener treatment processes utilizing supercritical CO_2, which boasts a lower critical temperature and reduced toxicity. Extensive research is required to thoroughly understand the precise effects of supercritical CO_2 on metal extraction, transport phenomena, and leaching kinetics properties. Integrating a supercritical CO_2 method with other approaches is essential for enhancing the sustainability of this separation process [40]. Supercritical fluids exhibit a heightened solvating capacity, low viscosity and surface tension, rendering them very effective for extractions. CO_2 stands out as a particularly suitable solvent for retrieving REEs, given its cost-effectiveness, relatively moderate critical pressure, and low critical temperature [77].

Lately, deep eutectic solvents (DESs) have emerged as environmentally friendly substitutes for traditional solvents owing to their affordability and reduced toxicity. These solvents are composed of hydrogen bond acceptors (e.g., quaternary ammonium salts) and hydrogen bond donors (e.g., carboxylic acids) [150]. Chen et al. developed a range of innovative Brønsted acidic deep eutectic solvents (DESs) derived from carboxylic acids and polyols, leveraging the substantial variation in metal oxide solubility within DESs. These solvents were employed for the separation of rare earth oxides, achieving purification of more than 99.9% [131].

While solvent extraction remains the primary commercial method for rare earth separation, its efficiency is constrained, often requiring multiple extractions to attain high purity. Moreover, many extractants pose risks due to their flammability and toxicity, leading to potential secondary pollution. Current solvent extraction research concentrates on enhancing separation efficiency and enrichment factors by optimizing selective extractants and organic solvents [151]. The current emphasis of solvent extraction research should prioritize the optimization of eco-friendly extractants with superior separation selectivity to enhance separation efficiency and enrichment factors. Additionally, there is a need for research into the recovery and reuse processes for the substantial volumes of organic solvent waste liquid generated by the existing procedures.

Adsorption

Recently, solvent extraction has become a popular method for recovering REEs from waste liquids, but its dependence on flammable and toxic organic solvents makes it unsuitable for industrial wastewater with low rare earth concentrations. In contrast, adsorption, which binds metal ions to an adsorbent, is more environmentally sustainable, reduces solvent and additive use, accommodates a wider concentration range, and is simpler. Given stringent environmental regulations, adsorption is superior commercially to sol-

vent extraction [72]. Adsorption emerges as particularly promising due to its simplicity, high extraction efficiency, environmental friendliness, and versatility for various applications [152]. An array of adsorbents, including activated carbon, carbon nanotubes, dried biomass, nanocomposites, polymeric materials, silica, and zeolites, have been explored for REE extraction from ores and WEEEs [153]. The adsorption of REEs is influenced by various mechanisms, each driven by specific forces. Chemical coordination involves forming complexes between the adsorbent and the REEs [46].

Numerous research groups have investigated bio-adsorbents, exploring natural materials like cellulose, brown marine organisms, and bacteria [154]. To enhance the efficacy of biological materials as adsorbents for REE separation and environmental purposes, biomass adsorbents are frequently combined with other substrates. A previous study utilized organophosphorus-modified metal–organic framework (MOF) derivatives, achieving adsorption of 37.2–57.5 mg/g of Er^{3+} at pH 5.5 [155]. Metal–organic framework (MOF) structures, combined with diglycolamide-based preconstructed resorcinarenes, have shown selective extraction of heavy REEs [46]. Li et al. observed a significant self-assembly phenomenon of tetrahedral RE4(L)4 cages in organic solvents [156].

In contrast to metal adsorbents, silicon-based materials exhibit lower selectivity to metals, mainly due to the characteristic of rare earth ions as Lewis acid ions. Functionalized adsorption materials containing groups with Lewis bases might provide advantages in rare earth separation processes [157]. Cui et al. [158] observed that REEs chemically adsorb onto DTPADA-modified silica (DTPADA SBA-15) through oxygen coordination at lower pH values. Surface complexation involves Van der Waals interactions or hydrogen bonding on the surface of the adsorbent [46]. Wang et al. employed wrinkled mesoporous silica (WMS) to produce wrinkled mesoporous carbon (WMC) with distinctive tapered wrinkles. They functionalized the carbon surface's oxygen-containing groups via acid treatment. This material demonstrated significant adsorption capacity and high selectivity for thorium (Th), facilitating the purification of rare earth elements from minerals [159].

Researchers created a GA-g-PAM nanocomposite by modifying natural polysaccharide gum arabic (GA) using a radial in situ graft copolymerization technique. This resulted in a significant increase in the specific surface area to 273.55 m^2/g, with a desorption reuse rate of up to three cycles, indicating its potential for rare earth adsorption and recovery from aqueous solutions [160]. Additionally, Gao et al. developed a phosphoric acid-functionalized mesoporous SBA-15 by grafting phosphite phosphorus oxychloride ($POCl^3$) onto SBA-15. This material maintained a 92.3% adsorption efficiency after three uses [161].

Carbon-based adsorbents, including mesoporous carbon, carbon nanotubes (CNTs), and graphene, are extensively employed in the separation of rare earth elements [83]. Graphene oxide (GO), possessing numerous oxygen-containing functional groups, demonstrates strong affinity and high adsorption capacity for rare earth elements. However, GO tends to aggregate and often necessitates functional modification or the introduction of other matrices to enhance its adsorption and separation capabilities. Incorporating nitrogen-containing groups can enhance selectivity for rare earth ions [72]. Zhao et al. improved GO by applying a one-step ultrasonic treatment to coat it with tris(4-aminophenyl) amine, resulting in a three-dimensional composite (GO-TAPA x) with an enlarged surface area and heightened selectivity for rare earth ions. This composite exhibited high stability and reusability in real water samples [162].

A bibliometric analysis revealed that carbon-based adsorbents constitute the subject of 17% of published articles. Silica-based chelating adsorbents represented 9%, while research on magnetic adsorbents (12%), MOFs (8%), and green adsorbents (11%) has witnessed recent growth. Furthermore, there has been a notable increase in studies on hybrid adsorbents integrating immobilized nanomaterials (Figure 6) [163].

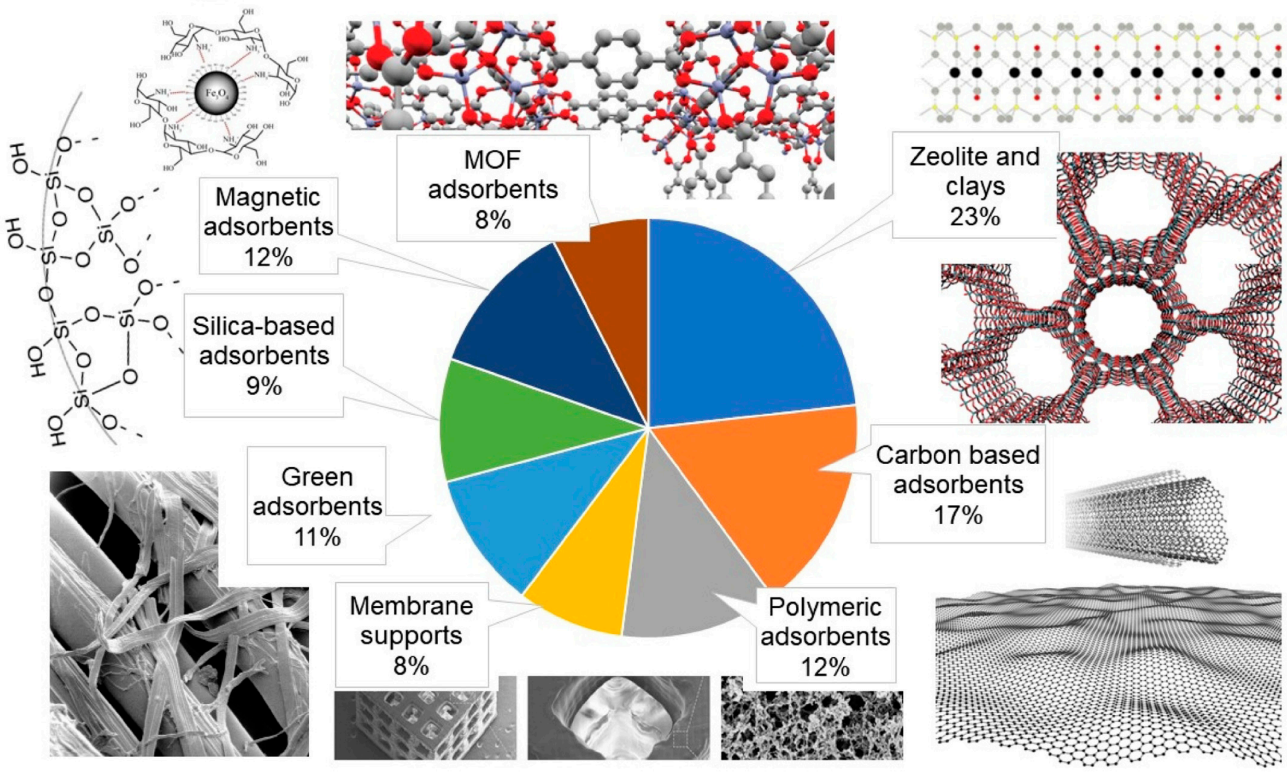


Figure 6. Adsorbents for the extraction and recovery of rare earth ions (Scopus database, 2017–2022). Reproduced with permission from Ref. [163].

In the context of REE biosorption, which involves both biological and non-biological sources, the interaction between ions and biomass encompasses various techniques such as absorption, adsorption, ion exchange, surface complexation, and precipitation [164]. Functional groups present in cell walls are instrumental in facilitating these interactions. Biosorption of REEs can be accomplished by both autotrophic and heterotrophic microorganisms, with studies [165] underscoring the importance of the cell wall in REE biosorption onto bacterial cell surfaces, [166,167]. For instance, Dong et al. investigated the adsorption of Tb and Dy using *B. subtilis* and found that the rare earth elements accumulated in the outer layers of the spores. Moreover, fungi have demonstrated effectiveness in recovering REEs from solutions [168].

Moreover, the inclusion of magnetic substances assists in regulating rare earth separation, recovery, and reuse [169]. Nanomaterial-based adsorbents exhibit heightened specific surface areas compared to conventional adsorbents, enhancing adsorption capacity. By incorporating acid-stable materials and grafting chelating agent groups onto the surface of silicon-based materials, their absorption and separation capabilities can be significantly improved, enhancing their performance. However, there may be a concern regarding the trapping of rare earth ions during regeneration and desorption potentially diminishing adsorption performance [72].

The adsorption separation of MOFs has led to exploration of the potential of COFs as a structured crystalline material for REE separation. Xiong et al. utilized a COF for separating REEs, thorium, and uranium [170], noting its high adsorption capacity and compliance with industrial separation standards for thorium. Nevertheless, further validation is required to confirm the capability of COFs to selectively separate REEs. To optimize the efficiency of biological materials as adsorbents, biomass adsorbents are frequently combined with other substrates. Despite the numerous benefits of bio-adsorbent materials, challenges persist in achieving the necessary concentration of adsorbents for industrial-scale use [72].

Membrane Separation

Membrane separation stands out for its efficiency, simplicity, energy-saving attributes, and ability to minimize organic solvent loss, rendering it a relatively eco-friendly technology for rare earth separation [81].

Due to its numerous benefits, including high selectivity and separation efficiency, as well as minimal energy consumption and waste generation, membrane separation techniques have emerged as a promising and environmentally friendly method for rare earth element separation. This is attributed to the diverse range of membranes available, offering various material compositions, porosities, and permeabilities, enabling the customization of conditions for REEs separation [171]. Over the past decade, reported membrane separation materials can be categorized into liquid and non-liquid (solid) membranes. The liquid membrane technique (LMT) comprises various extraction processes wherein the transportation of extracted species takes place through the membrane phase [172]. Liquid membrane extraction of rare earth metals stands out for its swift extraction kinetics, impressive enrichment ratio, integration of extraction with stripping, minimal reagent usage, and cost-effectiveness [173].

Liquid membranes encompass various types, including supported liquid membranes (SLMs), bulk liquid membranes (BLMs), hollow fiber-supported liquid membranes (HFSLMs), emulsion liquid membranes (ELMs), and electrostatic pseudo liquid films (ESPLMs), among others. In these membranes, lanthanide ions (Ln^{3+}) typically permeate from the feed solution through an organic phase to the stripping phase due to concentration gradients. Supported liquid membranes (SLMs), which consist of a porous support and a carrier for extracting target ions, have emerged as an effective system due to their high selectivity and reduced carrier consumption. SLMs are widely used in REE extraction. However, they have notable drawbacks such as low stability and blockage of support pores by aqueous solutions [172]. For example, J. Martínez et al. [174] utilized a robust flat SLM for recovering $Y3^{+}$-Nd^{3+}-Dy^{3+}, employing bis(2-ethylhexyl) hydrogen phosphate (D2EHPA) as an extractant and polymeric support. Nevertheless, the primary limitation of supported liquid membranes is the tendency of aqueous solutions to wet the porous structural support and inevitably block the carriers. Achmad et al. [111] investigated a solvent extraction process followed by a hollow fiber-supported liquid membrane (HFSLM), using a 1 mol/L D2EHPA extractant. The optimal recovery percentages achieved at 90 min for Nd^{3+}, Dy^{3+}, and Pr^{3+} were 63.13%, 15.21%, and 56.29%, respectively. Liquid membranes face several challenges, particularly regarding long-term utilization, stability, and extraction yield [171].

Non-liquid membrane processes involve carriers that are either chemically or physically bonded to the membrane or a porous supporting structure, thus allowing their utilization without the concern of extractant loss into the aqueous phase [172]. Polymer inclusion membrane (PIM) represents a relatively novel approach for the solid extraction of REEs, offering cost-effectiveness, high flux, and mechanical flexibility [175]. In a recent study focused on membrane stability, Tan et al. synthesized a nanoporous membrane from graphene wherein nitrogen doping as well as the pore size were controlled using a multi-confinement strategy. This material exhibited remarkable selectivity for the separation of individual REEs [176].

Presently, investigations into rare earth ion separation through membrane-based techniques have mainly concentrated on stable non-liquid membranes [72]. By employing solvent extraction followed by a hollow fiber-supported liquid membrane, retrieval rates of 15.21%, 63.13%, and 56.29% for Dy, Nd, and Pr, respectively, were attained from the leach liquor of waste permanent magnets within a duration of 90 min [111]. Siddiqui et al. (2021) evaluated the use of nanofiltration with a PVDF membrane to concentrate rare earth elements under varying operating pressures and different pHs. Optimal conditions resulted in a 90% REE recovery efficiency [177]. Zheng and colleagues employed a grafting-free technique, utilizing Dy(III) as the imprinting template. Through this method, they successfully produced six variants of imprinted membranes via evaporative self-assembly [178]. These

findings underscore the significant potential of imprinted membranes for the isolation of individual REEs.

It is worth noting that the separation of heavy rare earth ions poses significant challenges due to poor dissolution rates. Nonetheless, membrane separation methods are particularly suitable for addressing this challenge. Given the advancements in membrane separation technology, efforts should prioritize addressing issues such as membrane material swelling, as well as enhancing stability and recyclability. Increasing attention has been directed towards the separation and enrichment of REEs using the ELM technique [179]. However, despite its effectiveness, the commercial applications of this method are hindered by emulsion instability, which reduces extraction efficiencies [180]. Achieving the desired level of stability is crucial to overcoming this obstacle for industrial-scale application [181]. Since 1907, nanoparticles have shown potential for stabilizing emulsions due to their lower toxicity and cost, and they have been widely utilized in emulsification processes [182]. Lan et al. [183] investigated the stability of emulsions stabilized by silica nanoparticles and the cationic surfactant CTAB. Their findings demonstrated that silica particles can strongly adsorb to the oil-water interface with an appropriate addition of CTAB concentration. Zhou et al. [184] utilized Fe_3O_4 nanoparticles as stabilizers to prepare magnetic Pickering emulsions and found that Fe_3O_4 nanoparticles can stabilize dodecane–water and silicone–water emulsions. In recent years, efforts to address this issue have involved the investigation of multi-walled carbon nanotubes (MWCNTs) and Span-85, both individually and in combination, on membrane swelling and breakage. These studies, conducted by Payman Davoodi-Nasab et al., were conducted in batch mode, and dispersion Sauter mean diameter (D32) was compared to pure systems for continuous mode experiments. The results indicated that the simultaneous presence of MWCNTs and surfactant enhanced ELM stability, leading to a reduction in membrane swelling and breakage by about 46% and 63%, respectively [180]. Furthermore, compared to systems without MWCNTs, the decreased swelling, primarily attributed to the reduced amount of surfactant in the organic phase, resulted in diminished membrane breakage. Aziz et al. [185] employed organosilane-functionalized TiO_2 micro-flowers and nanorods to tune membrane surfaces for the desalination process via direct contact membrane distillation (DCMD). ZnO was also utilized to enhance membrane material properties and generate the required features, such as high stability and omniphobicity [186]. Despite membrane separation technology being underutilized in practical separation processes, its demonstrated effectiveness in selectively separating REEs underscores its potential [187].

Electrochemical Process

Beyond traditional methods for rare earth separation, new technologies like electrochemistry are revolutionizing the process with innovative approaches that are not only energy-efficient but also environmentally friendly. One notable example is a novel technique developed by Fang and colleagues, which uses a series of rare earth complexes featuring redox ligands. These complexes enable the electrokinetic separation, marking a significant advancement in the field [188].

Vapnik et al. devised a novel approach to synthesize a redox copolymer (Figure 7a) [189]. This polymer introduces an innovative method for the electrochemical regeneration of ion exchange copolymers. Gueroul et al., aiming to reclaim high-purity single REEs from neodymium–iron–boron magnets, also proposed a separation technology (Figure 7b) [95,163].

Kim et al. [86], discuss the electrorefining process, which employs techniques like electrodeposition, electrosorption, electrodialysis, and electrocoagulation, to separate target rare earth elements. Sanchez-Cupido et al. [190] used the electrodeposition of REEs. On-cathode oxide formation has been also extensively for the recovery of REEs. Electrosorption-charged rare earth elements ions migrate to the oppositely charged electrode and are separated from the liquid phase. Capacitive deionization (CDI) is a system that accumulates REEs in the electric double layer. While CDI primarily relies on electrostatic charges for separation and lacks high selectivity [191], its performance can be enhanced by incorporating a mem-

brane capacitive deionization. Electrochemical precipitation (EP) is utilized to enhance the recovery of REEs. In this light, Venkatesan et al. [192] introduced EPs to improve the recovery of Nd.

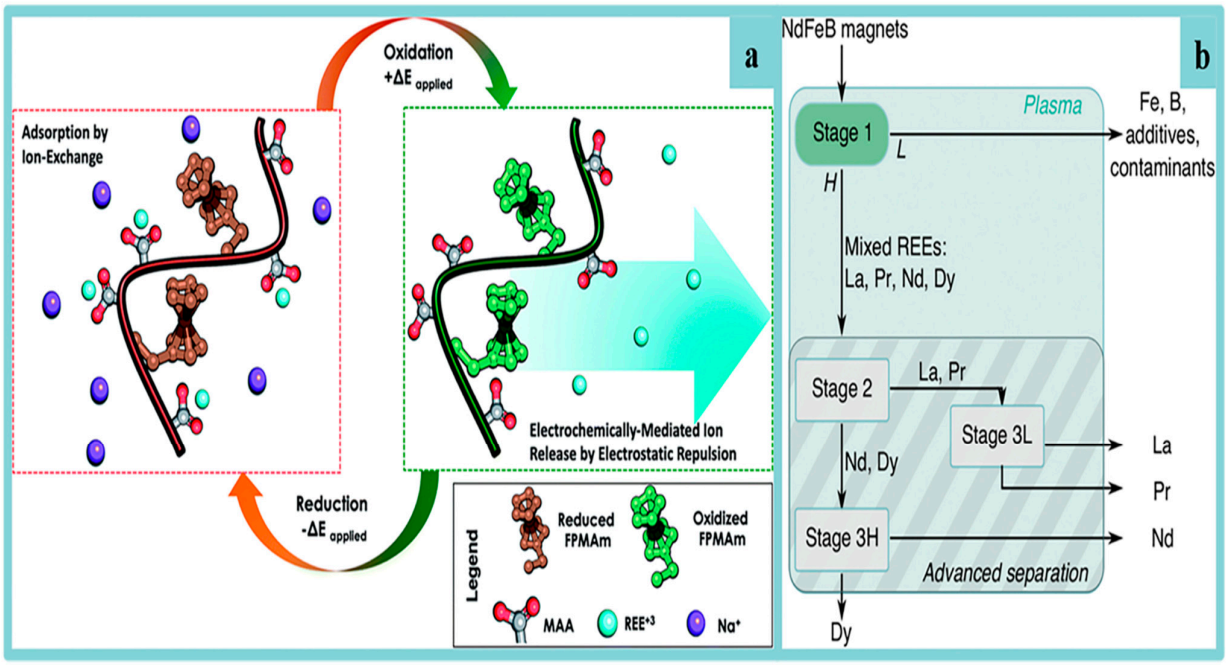


Figure 7. (**a**) Graphical presentation of reversible capture and release of REEs through electrochemical regeneration ion exchange [189]. (**b**) Graphical presentation of plasma recovery process for NdFeB magnets [95]. Reproduced with permission from Ref. [72].

This approach offers a novel avenue for metal separation. Electrochemistry introduces a fresh perspective for metal separation and investigation. Presently, electrochemical separation techniques have been employed for purifying individual rare earth elements owing to their environmental friendliness, minimal energy consumption, ease of operation, and continuous operation capabilities. However, if a metal necessitates being in a liquid state during this procedure, the electrolyte must exhibit a higher melting point than the metal, thereby restricting choices for both the electrolytic cell and the electrolyte selection.

Nowadays, the bioelectrochemical system (BES) emerges as a highly promising technology platform in effluent treatment. It is also gaining momentum for the retrieval of nutrients and REEs from liquid effluents [193]. Figure 8 illustrates a BES for recovering rare earth elements and other metals in various configurations.

In the context of recovering REEs using BESs, the electrons produced during the biodegradation of organic matter serve the purpose of reducing metals at the cathode of BESs. This reduction occurs because the redox potential of the half-cell reaction at the cathode is higher compared to the potential generated at the bioanode, resulting in a net positive potential for metal reduction. However, utilizing BESs for REE recovery from waste presents challenges. The effectiveness of biorecovery in BESs depends on the pH of the solutions, the potential of the bioanode, the redox potential, and the initial concentration and conductivity [193].

For example, Jin et al. employed a cylindrical turbulent cell with an enhanced surface area to improve mass transfer, resulting in approximately 93% copper extraction efficiency and 89.4% current efficiency [194]. In another study, an approach utilizing chloride-rich electrolyte was explored for the selective electrochemical recovery of tellurium from copper. This method led to a reduction in the copper reduction potential in an aqua regia-based solution due to the formation of monovalent copper chloride complexes. The highest tellurium recovery of 64% was achieved through a combination of electrodeposition, redox

replacement, and electrowinning processes [195]. Cobalt ions were effectively removed through capacitive deionization (CDI) using activated carbon cloth as the electrode material, resulting in a 36.5% removal efficiency with an initial cobalt concentration of 5 mg L^{-1}. However, competitive removal in the presence of strontium and cesium showed reduced performance for cobalt removal (8.4%), with adsorption capacities for Co, Sr, and Cs being 2.15, 2.19, and 1.84 mg g^{-1}, respectively [196].

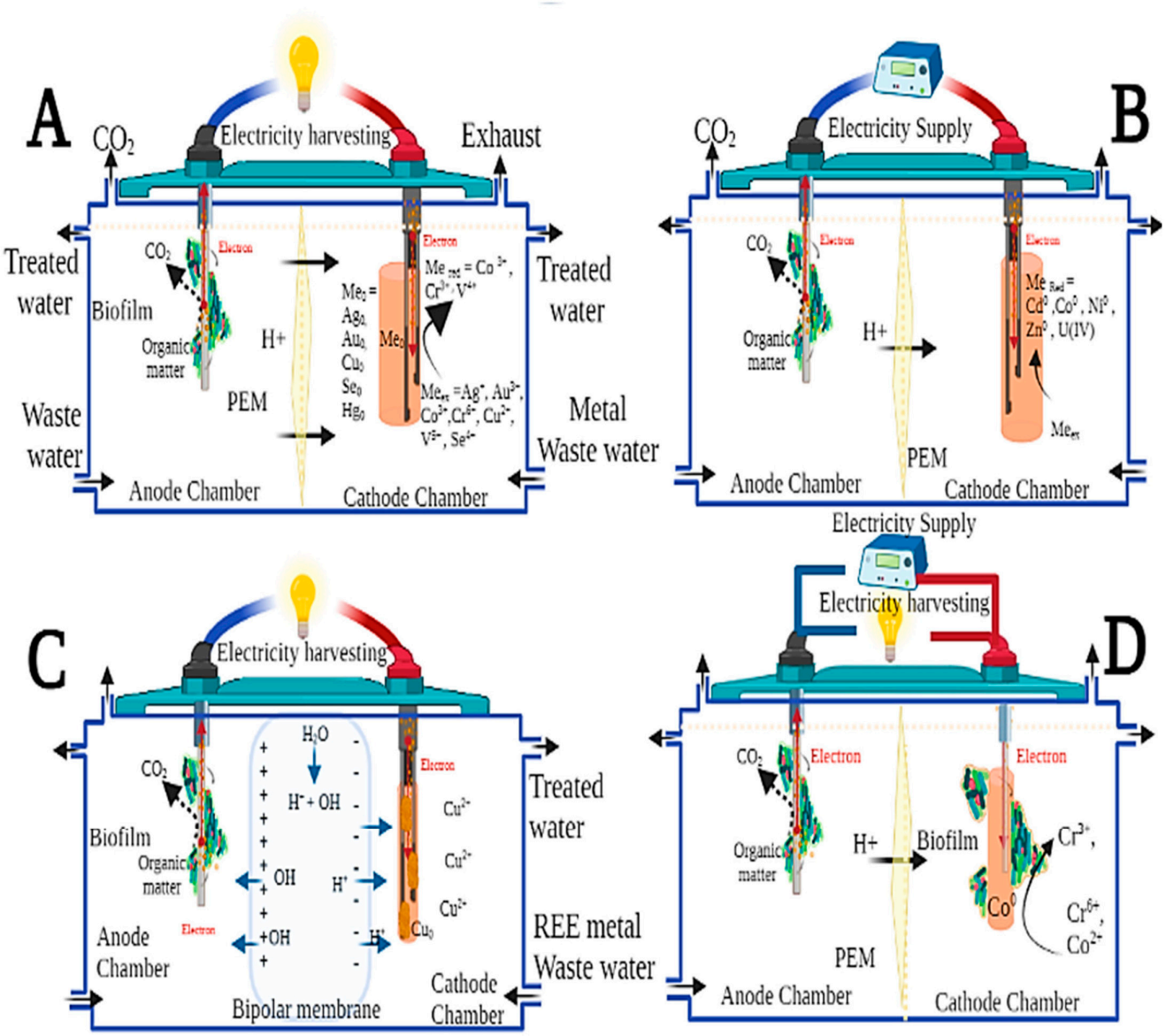


Figure 8. Extraction and retrieval of heavy metals (HMs) are facilitated through various configurations of microbial fuel cells and microbial electrolysis cells. These configurations include (**A**) microbial fuel cells, (**B**) electrolysis cells, (**C**) fuel cells with bipolar membranes, and (**D**) fuel cells and microbial electrochemical cells with biocathodes. Reproduced with permission from Ref. [46].

Selective separation of radionuclides cobalt and cesium from a solution was achieved using shock electrodialysis. This method creates a depletion zone that divides the system into a concentrated region and a deionized region [97]. However, there was a trade-off between cobalt removal and water recovery. For instance, the process removed 99.5% of cobalt while recovering 43% of the water. To enhance water recovery, the process could remove 98.5% of cobalt while recovering 66% of the water [197].

Therefore, prior to widespread adoption in industrial production, careful consideration must be given to selecting suitable materials for electrolytic cells and conducting thorough process research and development.

Bioleaching Process

Recovering rare earth elements (REEs) from leachate encounters difficulties with low recycling rates when using conventional hydrometallurgical and pyrometallurgical methods. Regardless of the approach, retrieving REEs from their primary source requires initial extraction and concentration through leaching methods, followed by separation techniques and purification to obtain a high-purity REEs solution [103]. Leaching, a critical step in this process, involves extracting REEs from the solid matrix using chemical leaching agents or microorganisms and their metabolic processes [198].

Natural biomaterials such as bacteria, algae, and fungi, as well as activated carbons and resins, exhibit the ability to adsorb REEs during recycling processes. While biomaterials generally have a lesser impact on the environment, they tend to be less efficient compared to chemical alternatives. Nonetheless, ongoing efforts to enhance the utilization of biomaterials include process optimizations and improvements in microbial strains through mutation technologies and other means. The quest for cleaner recovery strategies, therefore, necessitates a thorough assessment encompassing economic, technological, and environmental considerations [71]. In the majority of documented studies, secondary waste undergoes chemical and/or bioleaching [97]. Figure 9 depicts the chemical and biological leaching of REEs from phosphogypsum [46].

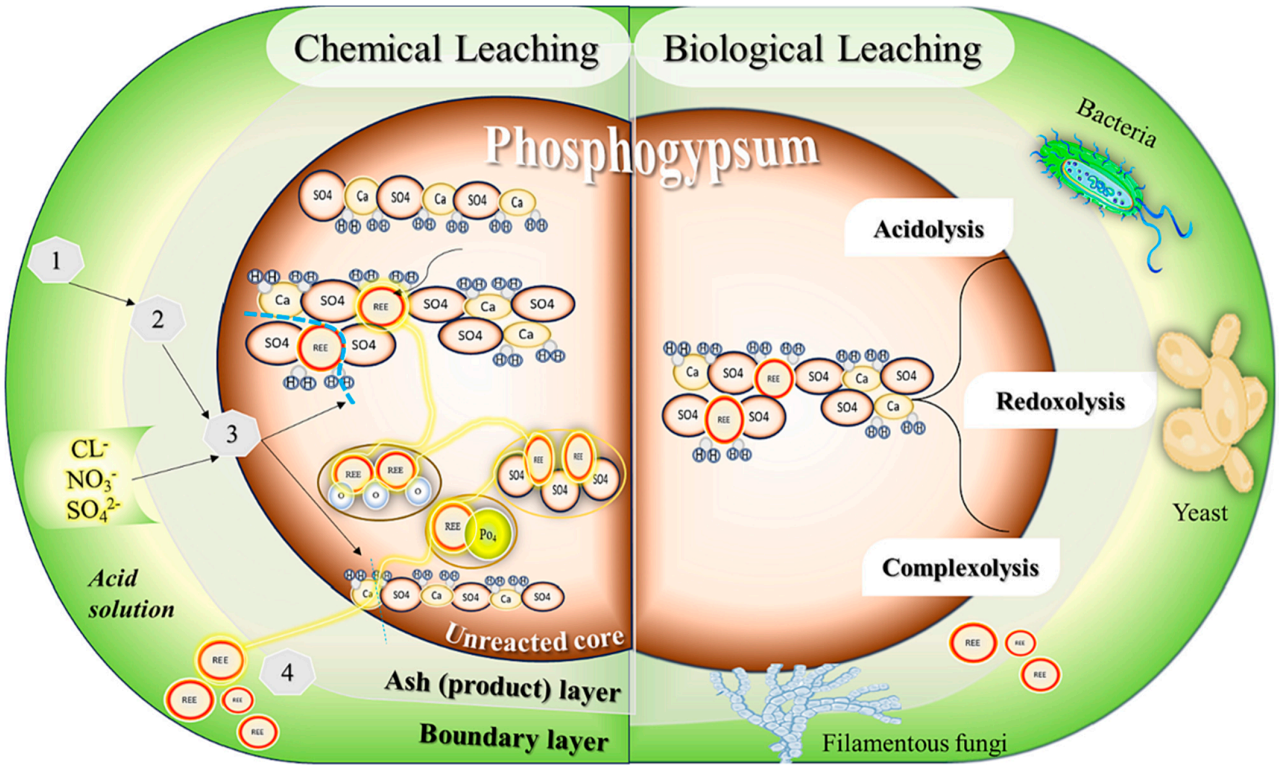


Figure 9. Chemical and bioleaching of REEs from phosphogypsum [46]. Microorganisms facilitate the dissolution of minerals and the extraction of rare earth elements (REEs) through several mechanisms. In acidolysis, microorganisms produce protons that interact with minerals, leading to their dissolution. Another mechanism, redoxolysis, involves microorganisms catalyzing the oxidation or reduction of minerals, resulting in the dissolution of REEs. Additionally, complexolysis occurs when microorganisms generate organic acids and other compounds that form complexes with REEs, aiding in their dissolution [46].

Utilizing acid leaching is a commonly employed method to extract REEs from diverse sources. Commonly used leaching agents include hydrochloric acid (HCl), nitric acid (HNO_3), sulfuric acid (H_2SO_4), aqua regia (3 HCl: HNO_3), hydrogen peroxide, cyanide, iodine, thiourea, halides, and thiosulphate, among others [91]. The effectiveness of acid leaching may vary due to numerous factors, including the acid type and concentration, the mineral composition of the source material, and operational variables. The initial stage involves leaching with an acid/base reagent (Equation (1)) [97].

$$RE_2O_3 + nH^+ \rightarrow nRE^{3+} + nH_2O \quad (1)$$

Sulfuric acid can be employed to convert rare earth element oxides into sulfates. (Equation (2)):

$$nRE^{3+} + nSO_4^{2-} + nH_2O \rightarrow UiRE_2(SO_4)_n \cdot H_2O \quad (2)$$

Alternatively, hydrochloric acid can be utilized to create chloride compounds (Equation (3)).

$$nRE^{3+} + nHCl \rightarrow nRECl_3 \quad (3)$$

However, acid leaching lacks selectivity, leading to the co-leaching of all impurities (such as Ca, Al, Fe, Mg, and Mn) along with REEs in solution. This poses a significant challenge for subsequent REE recovery, as well as for appropriate separation and purification processes. Additionally, recovering REEs at low concentrations and removing impurities (e.g., Ca and Al) proves to be both challenging and costly [199].

Acknowledging the drawbacks of traditional chemical leaching methods, researchers have turned their attention to alternative approaches. Biological leaching strategies have attracted significant interest. Bioleaching offers a potential solution to the limitations of conventional chemical techniques and represents a promising pathway towards a more environmentally sustainable future in REEs recovering [46]. Chelation of REEs from the solution: This eco-friendly approach employing microorganisms to solubilize REEs offers promise for enhanced recovery during the leaching stage. Bacterial strains under investigation for this purpose include *Acidithiobacillus ferrooxidans*, *Zygosaccharomyces lentus*, *Komagataeibacter hansenii*, and *Gluconobacter oxydans*. These bacteria typically facilitate REE solubilization through metabolic processes that involve organic acid production. Subsequent release of these organic acids into the environment promotes dissolution and researchers are now actively assessing the effectiveness of bioleaching for recovering REEs using both bacteria and fungi [62]. Bioleaching, for instance, has been proposed to offer a lower environmental footprint for both primary and secondary sources compared to many other REE recycling methods [200]. REE bioleaching can involve both autotrophic and heterotrophic microorganisms, offering adaptability to enhance REE extraction. Leaching, including bioleaching, plays a vital role by concentrating these elements in the solution. This process facilitates their recovery and separation involving various physicochemical and biological methods [46].

The use of bioreactors enables the manipulation of diverse operational parameters, facilitating the adjustment of REE bioleaching conditions according to factors such as the substrate, selected microorganisms, and their respective mechanisms. A range of bioreactor types, including column bioreactors, airlift percolators, batch reactors, and continuously stirred tank reactors, can be utilized for REE bioleaching. However, research on REE bioleaching remains relatively scarce, despite existing studies demonstrating varied extraction efficiencies [46]. In a particular investigation, circulating the leaching solution 35 times via the column led to the recovery of 67% of REEs from a gibbsite ore [201]. In continuous stirred tank bioleaching by *A. niger*, rare earth element leaching efficiencies varied between 27% and 62% [202]. Table 3 provides an overview of the recycling mechanisms of REEs and the main findings achieved through various methods.

Table 3. Recycling of REEs using a variety of methods.

Targeted Rees	Main Process	Subsequent Process	Key Findings	Reference
Eu and Y	Hydrometallurgical method	Oxidative leaching	The leaching process was observed to be markedly affected by temperature. Under the optimal leaching conditions, the efficiencies for both were up to 99%.	[203]
Eu and Tb	Hydrometallurgical method	Adsorption	Silica-based adsorbent with covalently immobilized fragments of 2,6-pyridinedicarboxylic acid (SiO_2-DPA) exhibits the ability for the quantitative adsorption with a pH greater than 1.7. Adsorption capacity was maintained over 97% after 10 iterations. The adsorbent recovered more than 80% of all REEs (excluding La).	[204]
Nd	Hydrometallurgical method	Ionic liquid	In the process of separating rare earths and transition metals from NiMH batteries, the recovery efficiency and purity of REEs can still surpass 96.4% and 99.8 wt%, respectively. This is achieved through the use of NaCl and Na_2CO_3 for the stripping process.	[109]
Ce, La, Tb	Hydrometallurgical process	Combination of pyrometallurgical and hydrometallurgical process; thermal treatment, leaching, precipitation, calcination	The efficiency of leaching with HCl surpassed that of leaching with H_2SO_4. In the suggested process, global recovery efficiencies of 82% (Ce), 75% (La), and 82% (Tb) were attained.	[205]
Y, Er	Bio-oxidation	*Komagataeibacter xylinus*, *Lactobacillus casei*	In each experimental scenario, yttrium and europium were selectively dissolved from fluorescent phosphor. Nevertheless, the outlined processes are currently inefficient and costly, necessitating optimization.	[206]
Y, La, Ce, Pr, Nd, Sm, Gd, Er, Yb	Biometallurgical process	Gluconate, ketogluconate	Gluconate consistently enhances higher leaching yields compared to ketogluconate under all examined pH conditions. The maximum leaching yields for REEs were achieved at pH = 3, resulting in the recovery of approximately 100% Mn, 90% Fe, 89% Co, 58% Ni, and 56% total REEs.	[200]
La, Ce, Pr, Nd	Superfluid extraction	CO_2 as a solvent	The efficiency of REE extraction was examined, and the optimal conditions were identified. Under the specified conditions, La, Ce, Pr, and Nd were recovered at rates of 86%, 86%, 88%, and 90%, respectively.	[207]

The effectiveness of REE recovery relies on various factors, including the technology utilized for extraction, the desired target elements for recovery, the types and origins of gangue minerals, the quantity of REEs present (ranging from milligrams to several grams), and the conditions (pH and temperature) of the extraction process. All REEs share the same external electronic configuration, with their ionic size being the primary distinguishing factor between them. Consequently, during compound formation, they do not directly engage in bonding with other elements. This similarity in physical and chemical properties among the elements poses challenges to separation processes and renders the REE recovery process more difficult [208]. The literature extensively documents the utilization of hydrometallurgical methods, likely due to their high recoveries (80–100%) and ease of control. In contrast, biometallurgical methods exhibit lower recoveries, and superfluid extraction yields comparable results [203]. More specifically, hydrometallurgical methods had recoveries of 99%, >97% [204] 96.4%, and 99.8 [109].

While both hydrometallurgical and pyrometallurgical processes are frequently utilized, each has specific drawbacks. For example, hydrometallurgical methods require significant amounts of water, causing a considerable environmental toll [86]. Nevertheless, hydrometallurgical processes have the capability to generate pure products from lower grade and intricate streams. The leaching process, involving fewer gas emissions, can be executed more straightforwardly and scaled up with lower operational costs compared to

the pyrometallurgical route [209]. Pyrometallurgical approaches involve high-temperature processes that demand significant energy inputs [86]. Despite its fundamental nature, the pyrometallurgical process faces a significant challenge in controlling gas emissions [71]. These limitations underscore the necessity to improve the efficiency, sustainability, and eco-friendliness of REE recovery methods [86]. Moreover, novel techniques such as biometallurgy, leveraging microorganisms for leaching and recovering REEs, present promising alternatives [210].

4. Occupational Safety

Human exposure to REEs stems from anthropogenic activities through various pathways. REEs become concentrated in atmospheric particulates due to human sources and are dispersed by strong winds. Workers in recycling facilities face significant exposure to REEs through inhalation, ingestion, and skin contact. Furthermore, REEs can enter the human body via the food chain, as they serve as feed additives for animal health and production. Additionally, they are present in pesticides, herbicides, and fertilizers to improve crop yield and quality [211]. As the use of REEs has become more popular, concerns about industrial health have grown. Large-scale applications over the past decades have resulted in widespread distribution of REEs, posing potential health risks to both humans and wildlife [212].

The health impacts of REEs on humans are still not well understood. In vitro and in vivo studies have shown various toxic effects of REEs, including neurological disorders, renal and lung fibrosis, pneumoconiosis, male infertility, and organ damage [212]. Epidemiological evidence also suggests that REEs adversely affect multiple body systems, including the central and peripheral nervous systems, respiratory system, and cardiovascular system [213].

According to Quiao et al., human exposure to rare earth elements (REEs) and the associated health impacts, particularly in occupational populations, remain insufficiently characterized. To address this, they aimed to investigate occupational exposure to REEs in a major REE processing plant in North China by assessing both external and internal exposure in workers [212]. Although there were significant differences in air concentrations between the control and exposure groups, the differences in REE levels in workers' blood were less pronounced. This might be due to the use of personal protective equipment, including masks, protective suits, and goggles, which minimized exposure in the occupational environment. The compositional profiles of REEs in both blood and air particles showed similarities between the exposure and control groups, suggesting the impact of mining and processing activities on the surrounding areas. External exposure in the workplace environment notably affected the internal exposure of workers to REEs. Moreover, sociodemographic and occupational factors, such as duration of residence and job type, could also influence the occupational exposure to specific REEs [212].

Exposure to rare earth elements (REEs), particularly Nd, has been associated with fingertip injuries due to strong magnetic forces and biocompatibility issues in medical applications. Injection of Nd compounds has led to symptoms such as fever, chills, abdominal pain, and blood clotting abnormalities. Nd dust exposure can cause lung embolism and liver damage. Dy, Y, Eu, Er, and Yb dusts are hazardous, with Dy being prone to ignition and producing flammable hydrogen gas when in contact with water. Ce exposure can result in itching, skin lesions, and heat sensitivity. Lu nitrate can burn or explode when heated, and Sc gas mixed with other compounds poses risks in workplaces like metal halide lamp manufacturing. Y compounds can cause respiratory symptoms. REE oxides and fluorides, including La, have induced pneumoconiosis in workers handling carbon arc lamps. Lu accumulation in organs has been reported, while Ho and Tm compounds may affect human metabolism, although their effects are debated [214,215]. Dermal fibrosis (nephrogenic systemic fibrosis) has been associated with gadolinium (Gd) used as a contrast agent in magnetic resonance imaging (MRI) [216]. A recent investigation revealed that gadolinium (Gd) contrast agents employed in MRI scans might pose a risk of severe kidney disease.

The study explored how Gd could cross the blood–brain barrier, accumulating in brain tissue, and examined its distribution and chemical interactions within the brain [217].

Additionally, preventing occupational diseases requires periodic health monitoring of workers [218]. An investigation of miners from Baiyunebo exposed to REEs indicated a significant reduction in bone mineral density, compared to the control group. This implies that prolonged environmental and occupational exposure leads to the accumulation of REEs, causing disorders in bone metabolism and a subsequent decline in bone mineral density [219]. Additionally, studies suggest a correlation between maternal exposure to REEs during pregnancy and increased occurrences of premature rupture of membranes, decreased neonatal hormone levels, and neural tube defects [220,221].

Regarding occupational safety and health concerns, exposure to REEs through short-term inhalation has been linked to acute irritant bronchitis, while prolonged exposure has been associated with pneumoconiosis and progressive pulmonary fibrosis. The debate over whether interstitial pulmonary fibrosis results from REEs or radioactive contaminants arises, as REE compounds might cause benign pneumoconiosis, with uncertainty about its progression after exposure termination [37]. Elevated REE levels in miners' hair have shown correlations with Fe and Ca, coupled with reduced bone mineral density at various bone sites [219]. Elevated levels of REEs in the bloodstream have been linked with significant DNA oxidative damage and an elevated blood concentration of chromium in workers exposed to chromate [222]. The urine and blood of e-waste recyclers contain higher levels of Eu, with this observation specifically applying to urine in the case of La [223]. REEs have been detected in triple-negative breast cancer cells in a ceramic industry female worker [224]. Table 4 presents occupational and health safety issues associated with REEs.

Table 4. Occupational and health safety issues with REEs [215].

Symbol	Name	Occupational Health and Safety Concerns
Sc	Scandium	The primary concern lies in the workplace, where inhaling vapors and gases can occur.
Y	Yttrium	Exposure to yttrium compounds by workers can result in symptoms such as shortness of breath, coughing, chest pain, and cyanosis. Yttrium dust is flammable.
La	Lanthanum	The use of lanthanum in carbon arc lights has exposed individuals to rare earth oxides and fluorides, occasionally resulting in pneumoconiosis.
Ce	Cerium	Exposure to cerium may cause itching, heat-sensitivity and skin lesions.
Pr	Praseodymium	Praseodymium compounds are subjects of controversy regarding their biological roles.
Nd	Neodymium	Inhaling neodymium dust can result in lung embolisms, and prolonged exposure may harm the liver. Challenges related to biocompatibility have limited the widespread adoption of neodymium magnets. Improper handling can lead to swift and forceful attraction, posing a risk of injuries.
Pm	Promethium	It has no biological role. Sealed promethium-147 is not hazardous, but if the exterior is damaged, it becomes hazardous to human health and the environment.
Sm	Samarium	The biological role of samarium metal compounds in the human body is a controversial topic.
Eu	Europium	Dust of its metal compounds poses fire and explosion hazard.
Gd	Gadolinium	Gadolinium's biological roles are not extensively documented, but its compounds serve as research tools in biomedicine.
Tb	Terbium	Terbium compounds and their roles are a controversial subject.
Dy	Dysprosium	Dysprosium powder can pose an explosion risk. Thin foils of the substance can be ignited by sparks or static electricity. Such fires cannot be extinguished with water and may react with it, leading to the creation of flammable hydrogen gas.
Ho	Holmium	Holmium compounds are a controversial subject regarding their biological roles in humans, but may be able to stimulate metabolism.
Er	Erbium	Metallic erbium dust poses a fire and explosion hazard.
Tm	Thulium	Thulium compounds are a controversial subject in terms of their biological roles but they might stimulate metabolism.
Yb	Ytterbium	Despite its relatively stable chemical nature, ytterbium should be stored in air-tight containers to shield it from air and moisture. Metallic ytterbium dust presents a risk of fire and explosion.
Lu	Lutetium	Lutetium nitrate has the potential to explode and burn when heated. Lutetium has no known biological role, but it concentrates in bones, liver, and kidneys.

Ensuring workplace safety during the REE smelting and recycling process requires thorough management of hazardous substances, appropriate protective equipment, and regular safety education. This is crucial due to the potential hazards like nitrogen oxides or chlorine gas from the use of nitric acid or aqua regia. Effective monitoring systems

should adhere to relevant regulations, standards, or legislation from industries dealing with similar processes. Establishing exposure limits for REEs requires a comprehensive hazard/risk assessment, complemented by on-site field surveys to verify workplace exposure conditions.

Various environmental concerns arise from mining activities, yet REE production introduces specific risks such as radioactive pollution and REE toxicity, garnering increasing attention. Although radioactive pollution has long been recognized as a significant public health issue, the direct toxicity of REEs is frequently overlooked due to their naturally low environmental presence and the agricultural benefits they offer [225]. Rare earth ores and concentrates inherently contain radioactive elements like thorium, uranium, and radium, which inevitably end up in tailings or smelting slag, elevating radiation levels above background levels [226]. Workers in the REE mining and processing sector face chronic exposure to hazardous substances through ingestion, inhalation, and skin contact, potentially leading to organ or tissue damage over time due to bioaccumulation. Radiation is typically considered the primary health threat for workers in REE mining areas [225]. Thorium, particularly hazardous due to its natural decay, can enter the body as insoluble dust, accumulating in the lungs and causing direct damage [227]. Proper management of the radioactive fraction in REE production is crucial for environmental sustainability and human health. Actinides like uranium and thorium, sharing chemical properties with REEs, are often co-dissolved in current hydrometallurgical processes, leading to radioactive contamination throughout the extraction cycle [228].

5. Future Perspectives and Conclusions

The increasing demand for REEs, propelled by diverse sectors, presents a significant opportunity for the recycling industry to reclaim these valuable resources. It is crucial to recognize that recycling is just one of several potential responses to perceived risks in REE supply. The increased emphasis on recycling REEs shows potential in addressing key issues, including meeting growing demand, improving supply security, and resolving the balance problem between REEs with higher and lower demand. The future outlook for REE recycling depends on the specific material being recycled.

The recycling and reclamation of rare earth elements (REEs) hold the potential to generate value chains and employment opportunities while mitigating the environmental and health risks associated with Waste Electrical and Electronic Equipment (WEEE) [229]. Scaling up REE recycling could address critical issues related to these elements, such as meeting growing demand, enhancing supply security, and reconciling the imbalance between high and low demand REEs and their availability from primary mined sources [18]. Evaluation at pilot scale is essential to identify extraction methods with optimal recovery rates and minimal costs. Such pilot-scale assessments should replicate industrially relevant conditions, including typical WEEE compositions, which are often overlooked in laboratory-scale investigations [16].

Adsorption is suitable for treating low-concentration rare earth solutions as it can concentrate rare earth ions. However, achieving both selectivity and high adsorption capacity remains a challenge for adsorption materials. Current research in this field is still in the experimental phase and has not undergone large-scale production testing [72]. Electrochemical processes provide an alternative to reduce chemical consumption and wastewater generation, especially when integrated with other techniques, resulting in high-purity rare earth products. Carbon-based materials offer another alternative to enhance product purity [40]. Electrochemical separation offers advantages making it suitable for large-scale industrial REE extraction from minerals or compounds. However research on the separation of individual REEs is still limited, requiring substantial experimental data to verify its effectiveness. Overall, there is significant potential for further improving REE separation technologies and enhancing separation efficiency [72].

Data on the environmental and climate impacts of REE recovery from WEEE are scarce. Thus, conducting a comparative Life Cycle Analysis (LCA) of REE recovery from

WEEE versus conventional rare earth ores is imperative. Unlike studies focusing on individual process components in the value chain, a LCA provides a holistic understanding of the climate and environmental footprints of a process or product across the entire value chain [230].

While recycling strategies pose challenges, they also present great opportunities for their sustainable management. Hydro- and pyrometallurgy are widely applied technologies, but concerns such as energy and chemical consumption, as well as their emissions, require further research. Greener alternatives, including bio-based processes, show promise [40]. However, the current state of REE recovery is not sustainable [40]. The selection of a technology for economic feasibility of the recovery process requires taking various parameters into consideration, such as investment expenses, labor and operational costs, REE remnants in the waste, recovery efficiency, and environmental impacts.

Interdisciplinary research efforts spanning technology, environment, and economics are essential for developing innovative, eco-friendly, and cost-efficient methods for extracting REEs. The wide range of final applications poses challenges for the extraction and reuse of REEs. Despite this complexity, certain rare materials exhibit high recycling potential, proving economically beneficial [38]. Although research has been conducted to identify substitutes for REEs in various applications, breakthroughs are yet to occur, requiring further research.

Researchers are also exploring circular economy strategies, aiming to understand how recycling, recovering, or reusing these elements can be adopted. For the operation to be financially viable, operational costs must be low, as energy inputs are a major cost driver with adverse environmental effects. Comparative assessments of REE recovery and costs for various techniques are needed, and solutions with the best recovery and lowest cost should undergo pilot-scale evaluations to improve the design and operating parameters.

In terms of occupational health, additional research is required to evaluate hazards or risks for workers, including determining REE quantities at which health issues may arise. Toxicology testing of REEs calls for the establishment of standardized protocols to facilitate comparison and explanation of data derived from this research [30,37,215].

Author Contributions: Methodology, D.A.G., M.C. and G.Z.K.; writing—original draft preparation, D.A.G., M.C. and G.Z.K.; writing—review and editing, D.A.G., M.C. and G.Z.K.; supervision, G.Z.K. All authors have read and agreed to the published version of the manuscript.

Funding: We acknowledge support of this work by the project "Advanced Nanostructured Materials for Sustainable Growth: Green Energy Production/Storage, Energy Saving and Environmental Remediation" (TAEDR-0535821) which is implemented under the action "Flagship actions in interdisciplinary scientific fields with a special focus on the productive fabric" (ID 16618), Greece 2.0—National Recovery and Resilience Fund and funded by European Union NextGenerationEU.

Data Availability Statement: All data analyzed during this study are included in this published article.

Conflicts of Interest: The authors declare no conflicts of interest.

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