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A comparative state-of-technology review and future directions for rare earth element separation

Emmanuel Ohene Opare [a,b], Ethan Struhs [a], Amin Mirkouei [a,c,*]

[a] Department of Mechanical Engineering, University of Idaho, Idaho Falls, ID, 83402, USA
[b] Department of Systems Science and Engineering, Idaho National Laboratory, Idaho Falls, ID, 83402, USA
[c] Industrial Technology and Technology Management Programs, University of Idaho, Idaho Falls, ID, 83402, USA

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A B S T R A C T

Growing consumption of rare earth elements (REEs) due to their critical roles in various sectors (e.g., healthcare, energy, transportation, and electronics) has gained attention and stimulated research efforts in industry and academic communities. This study provides an overview of the existing REE production and recovery pathways, identifies critical challenges of the current techniques, and highlights opportunities for multidisciplinary research to achieve more effective solutions. A comprehensive classification of REE separation techniques is presented through narrative and systematic literature reviews, including qualitative analysis and classic bibliometric techniques, to assess the usefulness of identified methodologies and approaches. It is found that the top three most explored and mature separation techniques in various phases (solid and liquid) between 2015 and 2020 are leaching, solvent extraction, and plasma; and the top three study fields are chemistry, engineering, and metallurgy. It is further found that the dominant REE separation technique across over 40 fields of research is the use of acids, bases, ionic liquids, and salts for leaching REEs. It is concluded that agromining approach, using hyperaccumulator plants capable of absorbing REEs through their roots and leaves, can be a practical approach for sustainable REEs recovery from secondary sources and end-of-life products, such as electronic devices.

1. Introduction

1.1. Motivation and challenges

Across the globe, there is a growing demand for advancing technologies made from rare earth elements (REEs), particularly consumer electronics (e.g., cellular phones and computer tablets) that is predicted to reach 2.5 trillion dollars by 2030 [1]. REEs (e.g., cerium and dysprosium) are necessary components of various advanced technologies (e.g., batteries, catalysts, and magnets) that play a crucial role in energy security, economic growth, and environmental sustainability [2]. Therefore, research and advancements toward sustainable REEs recovery from secondary sources are necessary to mitigate the environmental impacts and replenish the resources required to meet future technological needs [3–5].

The increasing global population, changing demographics, and improving business environments have stimulated research efforts in diversifying light or heavy REEs supply to address major techno-economic and environmental challenges (e.g., energy usage and hazardous conditions) associated with diverse recovery strategies [6,7]. In 2019, China supplied more than 80% of the world's REEs, thereby making China the dominant supplier of global REEs to date [8]. The enormous importance of REEs makes these essential elements susceptible to global supply risks, especially when REEs production comes from finite ore sources [9,10]. Sustainable approaches (e.g., urban mining and e-waste recycling) for recovering these critical metals (e.g., neodymium) from end-of-life products (e.g., computer hardware, speakers, semiconductors, compressors, and batteries [11]) due to the high concentration of REEs in consumer electronics, can be one of the front-runners to address the current and future market needs [12].

1.2. Background

REEs are defined as 15 lanthanides (chemical elements), which are

Abbreviations: Ce, Cerium; Dy, Dysprosium; Er, Erbium; Eu, Europium; Fe, iron; Gd, Gadolinium; Ho, Holmium; In, Indium; La, Lanthanum; Lu, Lutetium; Nd, Neodymium; Pr, Praseodymium; REE, rare-earth element; Sb, Antimony; Sc, Scandium; Sm, Samarium; Tb, Terbium; Tm, Thulium; Y, Yttrium; Yb, Ytterbium.

* Corresponding author. Department of Mechanical Engineering, University of Idaho, Idaho Falls, ID, 83402, USA.
E-mail address: amirkouei@uidaho.edu (A. Mirkouei).

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cerium (Ce), dysprosium (Dy), erbium (Er), europium (Eu), gadolinium (Gd), holmium (Ho), indium (In), lanthanum (La), lutetium (Lu), neodymium (Nd), praseodymium (Pr), samarium (Sm), terbium (Tb), thulium (Tm), ytterbium (Yb), along with scandium (Sc) and yttrium (Y), and occur together in nature and possess a similar trivalent oxidation state [1]. RREs are known as 'rare' not because they are exceptionally uncommon, but are not found in economically available deposits [13]. Due to their unique physical and chemical properties, REEs have become essential components to clean energy technologies, such as wind turbines, electric vehicles, high-efficiency lighting, batteries, and hydrogen storage [3,6,14]. Additionally, rare earth metals are increasingly used in colored phosphors, lasers, and high-intensity magnets [15]. For instance, phosphors are essential in lighting applications because of their exceptional qualities, including emitted color determination, high thermal stability, high efficiency, excellent luminous intensity, low energy consumption, and durability [16]. Phosphors containing REE ions are preferred to use in lighting applications because they can emit various colors of the visible light spectrum and be environmentally friendly and non-toxic [17]. According to the recent studies (e.g., United States Geological Survey, Mineral Commodity Summaries 2020 report), the estimated REE distribution by end-use are catalyst (~75%), ceramics, glass, and polishing (~10%), metallurgical applications and alloys (~5%), and other (~10%), as shown in Fig. 1 [18,19].

Over the past two decades, the need for a steady and unhindered supply of REEs from diverse sources has arisen to satisfy global needs [20]. Predominant sources of unrefined light REE minerals are bastnaesite, monazite, xenotime, fergusonite, loparite, apatite, and kaolinite [21,22]. Bastnaesite (or bastnäsite, CO_3F) holds the highest concentration of light REEs, which is the primary source of Ce, La, Nd, and Pr [2]. Currently, China has the largest deposits of bastnaesite and produces 78% of global rare earth oxide and REEs [9,18]. Overcoming light REE mining challenges could be solved with efficient REE recovery and separation techniques among end-of-life products [23,24]. Recent studies reported that large concentrations of REEs are available in landfills [25]. According to the United Nations' studies in 2019, the world produces 50 million tons of e-waste (e.g., used TVs, computers, monitors, and cellphones) per year, which is worth over $60 billion annually, and only 20% of them is recycled [26]. They reported that there is over 100 times more gold in a ton of e-waste than a ton of gold ore, and they predicted global e-waste would reach 120 million tons by 2050 [26]. Additionally, prior studies reported that deposits of platinum group metals (e.g., ruthenium, rhodium, palladium, osmium, iridium, and platinum), gold, and silver in e-waste deposits at landfills are on average similar in concentration to large ore deposits currently available from traditional mines [11].

E-waste recycling practices include manually grinding consumer electronics (e.g., circuit boards) into fine powders for exporting, and open incineration of cables and plastic to liberate metals, such as copper [27]. In cathode ray tubes, the copper coil is stripped off, and the glass is crushed manually with stones. These practices are hazardous and damage or destroy REEs that could be harvested. With continued interest in clean energy technologies coupled with projected growth in power and transportation across the globe, the need for integrated techniques, such as the use of ligands (molecular recognition technology) for REE production must increase proportionately [28]. The existing REE production approaches can be classified into four main techniques: extraction/pre-concentration, purification, separation, and refining (Table 1).

Extraction and pre-concentration technique.

Earlier studies employed various technologies (e.g., adsorption, ion floatation, and precipitation flotation) for REEs extraction and pre-concentration [29–31]. Hu et al. (2016) investigated pre-concentration techniques for the extraction of REEs, such as Nd and Dy, using a solid-phase microextraction method and concluded that the solid-phase method was preferred over liquid-liquid extraction of REEs for its robustness and easy handling [32]. Hosseinzadegan et al. (2016) investigated the use of coated bioparticles with an ionic liquid to pre-concentrate REEs after leaf samples (tea leaf) were dissolved in water, using a microwave oven [33]. Their study resulted that the

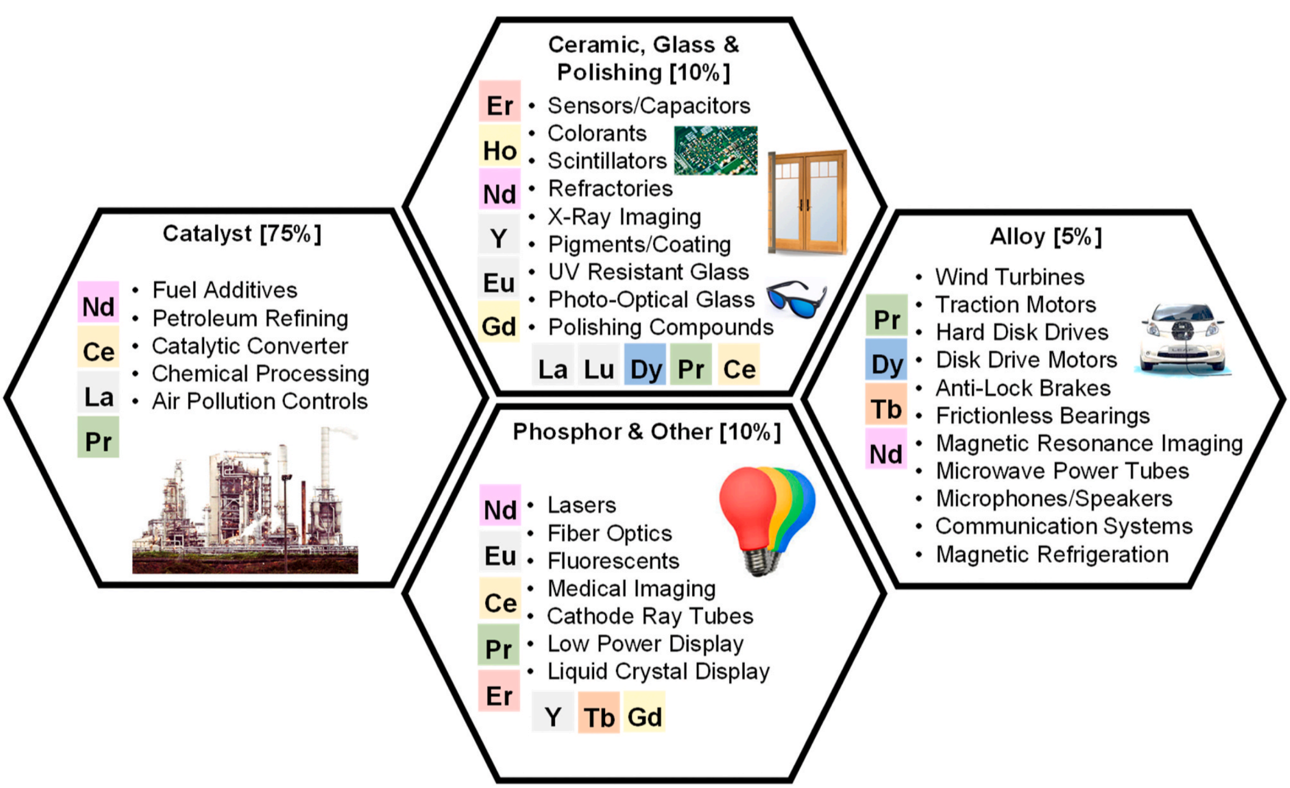

Fig. 1. Applications of REEs in various sectors [18,19].

Table 1
Current REE production approaches.

Technique	Chemical	Physical/Thermal	Electrical	Biological/Agronomical
Extraction/Pre-concentration	• Solid-phase extraction • Liquid-liquid extraction • Precipitation flotation • Liquid-phase extraction • Adsorption • Coated magnetic nanoparticles with ionic liquids • Dissolved air flotation	• Reverse osmosis • Microreactors • Magnetic nanoparticle adsorption • Ionic imprinted polymers • Silica sorbents • Nanofiber adsorbents	• Electrode ionization	• Ionic liquid coated bioparticles
Purification	• Selective oxidation/reduction • Fractional precipitation • Fractional crystallization • Vaporization • Ion exchange resin • Reduction-distillation • Membrane electrolysis • Molecular recognition (ligands)	–	• Electrocoagulation/ flocculation	–
Separation	• Leaching (ionic liquids, acids, bases, and salts) extraction • Liquid and gas chromatography • Sorbent membrane separation • Chemical coagulation • Photochemistry • Ion floatation • Solvent extraction	• Microwave separation • Membrane distillation • Microfluidic devices • Plasma separation • Forward Osmosis • Ultrasonic leaching • Nanofiltration	• Electrochemical separation • Electrostatic separation • Electrodialysis • Eddy current	• Adsorption • Bioleaching • Phytomining/Agrominig (using shoots, leaves, roots, and mushrooms) • Biosorption
Refining	–	• Vacuum reduction-distillation • Silocotheric/ methalothermic reduction	• Electrorefining • Electrowinning • Solid-state electron transport	–

dispersed particle extraction method eliminated plasma loading challenges, and attributed to solid-phase extraction of REEs. Also, they concentrated 50 ng per gram from the tea samples used and concluded that natural bioparticles coated with ionic liquid could be used for extracting and pre-concentrating REEs [33]. Recently, Lerner et al. (2019) studied the use of a chemically modified silica-gel as an ion-exchange resin for pre-concentrating REEs [34]. Their results show that the resin had the desired properties for pre-concentrating REEs and could be used in the future to extract actinides and lanthanides from very dilute aqueous solutions [34]. More recently, Wang et al. (2020) investigated the use of amine sorbents for the selective recovery of Dy and Yb from aqueous solutions [35]. Their results show that the sorbent could be used for pre-concentrating 7.6 wt% (weight of solute/weight of solvent) REEs in solution, which exceeds the United States Department of Energy REEs pre-concentration objectives. Additionally, the sorbent allowed the cyclic recovery of over 80% of parts per billion of REEs and around 0% of the parts per million of sodium, magnesium, and calcium from an acid mine drainage solution. This achievement highlights the applicability of the sorbent for pre-concentrating REEs from practical waste streams [35]. Further details on REEs extraction/pre-concentration techniques have been provided by Pereao et al. (2018) and He et al. (2019) [36,37].

Purification techniques.

Several studies employed various techniques (e.g., vaporization, reduction-distillation, and membrane electrolysis) for REEs decontamination and purification [38,39]. Royen et al. (2016) investigated REE purification techniques (e.g., selective oxidation/reduction, fractional crystallization, and fractional precipitation), using chemical reduction, salts, and the small difference in the basicity of REEs to selectively decontaminate or purify them [38]. Recently, El Afifi et al. (2019) conducted a study concerning the efficient removal of radionuclides and iron in REE liquor from monazite ore and observed that an admixture of sulfate-sulfide solution (0.058/0.04 mol/L) was used, the average percent removal of undesired species reached 96%. When a potassium iodate solution of 0.155 mol/L was used, the average percent removal of impurities was 99% [40]. They concluded that iodate solution is an efficient and selective agent for the removal of radium isotopes, Pb-210, Th(IV), and Fe(III) from rare-earth chloride liquor without loss in lanthanides that these reagents eliminate the human risks associated with radionuclides and achieve highly purified REEs from monazite ore. More recently, Silva et al. (2020) conducted a study on the effects of different neutralization reagents to selectively remove impurities in REE sulfuric liquor and concluded that the choice for the best purification condition was achieved when the pH of limestone pulp was initially increased to 3.5, then followed by adding the lime pulp to the mixture to achieve a pH of 5.0. Under this condition, Fe^{3+}, PO_4^{3-}, and Th^{4+} ions were removed entirely, and Al^{3+}, UO_2^{2+}, and SO_4^{2-} concentrations were reduced by 99%, 87%, and 37% wt./wt., respectively, in the purified rare earth liquor. Furthermore, this method also resulted in the lowest REE losses (8.3% wt./wt.) and required a low reagent consumption (4.3 kg/m^3) while producing low residues (7.5 kg/m^3) [41]. More detailed information on other purification techniques has been provided by Wang and Cheng (2011) [39].

Separation techniques

Earlier studies employed various technologies (e.g., solvent extraction and liquid membranes) for REEs separation [42,43]. Makanyire et al. (2016) investigated the use of ionic liquids (a family of organic molten salts with low or negligible vapor pressure, which may be formed below 100 °C) for the elemental separation of REEs [44]. They also discussed the study by Yang et al. (1995), where an electrostatic pseudo-liquid membrane is used in pre-concentrating and separating Sc from other rare-earth and contaminants. Their results show that Sc could be separated at 96% purity in a single step [42,44]. Recently, Pereao et al. (2018) reported that polymer adsorbents could separate around 95% REEs when used [36]. Additionally, a hydrometallurgical process for the recovery of La, Y, and Gd from spent optical glass containing lanthanum oxide, yttrium oxide, and gadolinium oxide, using sodium hydroxide, then followed by hydrochloric acid leaching of the residual solids, achieved recovery and separation of 99% La, 100% Y, and 100% Gd from the glass [36]. Also, Meshram and Abhilash (2019) discussed various REE separation techniques and a variety of feedstocks for consideration [45]. More recently, da Costa et al. (2020) studied the recovery of rare earth metals from aqueous solutions by bioabsorption, using non-conventional materials [46]. They reported that acidic solutions containing various hydrochloric acid concentrations (ranging between 0.01 and 1.0 mol/L) could result in an increase in desorption efficiency of up to 99% in a single regeneration cycle [46]. Further details on separation techniques have been provided in the Narrative Literature Review (Section 2).

Refining techniques

Earlier studies employed various technologies (e.g., electrowinning, electrorefining, and solid-state electro-transport) for ultra-purification of rare earth metals (e.g., Gd and Nd), using solid-state electro-transport technology [47,48]. Fort et al. (1987) achieved approximately 99.94% purity for Gd and 99.97% for Nd, and expected to reach 99.99% overall purity for other REEs [48]. Tunsu et al. (2015) studied air-jetting hydrometallurgical technique and reported that air-jetting could be used on products containing phosphors, particularly from the bulb holder of a lamp, to permit the recovery of phosphors for downstream refining [49]. They achieved around 95% recovery efficiency in the first step. In the second step, an air classifying machine achieved over 61% and 67% recovery of red phosphors and green phosphors, respectively. They reported that the air classification method achieved up to 95% purity for red phosphors. Compared to other solvent extraction processes where chemical solvent extractants (e.g., Di-(2-ethylhexyl) phosphoric acid, 2-ethylhexylphosphonic acid mono-2-ethylhexyl ester, Aliquat 336, and Tributyl phosphate) diluted in kerosene are used, each extraction stage is coupled with two to four scrubbing and stripping units to achieve purity levels greater than 99% for individual REEs [49]. More recently, Yang et al. (2020) conducted a study on the recovery of REEs from rare earth permanent magnets (e.g., Neodymium–Iron–Boron), using an electrorefining technique in a molten fluoride electrolyte [50]. They reported that the separation rates of Nd and Pr improved in the molten fluoride electrolyte with increasing current. The separated rare-earth ions were drawn to the cathode by electrolysis, leaving the porous Fe_2B alloy and metallic iron (Fe) in solution. More details on refining approaches have been provided by Meshram and Abhilash (2019), and Yang et al. (2020) [45,50].

1.3. Objective and scope

The recycling rate of REE is low, and substitutes in most cases are either inferior or still undiscovered [2]. The reliance on REE mining from ore does not provide an environmentally sustainable solution and requires significant resources (e.g., materials and energy), as well as water and land use, while generating large amounts of air emissions and solid waste. The primary focus of this study is to investigate REE recovery methods and to achieve energetically and environmentally sustainable techniques for the separation of REEs into the light and heavy REEs, especially individual oxides (e.g., Nd, Dy, and Pr). The specific objectives of this study are to (1) investigate the current state-of-the-science and gain an in-depth understanding of intricacies, (2) identify the chronological evolution of the current and developing REE separation techniques, and (3) highlights the challenges and potential future directions for sustainable REE recovery. The literature review conducted in this study includes both narrative and systematic reviews to assess the prior developed methods and approaches and identify high potential technologies for REE separation from end-of-life products and secondary resources.

2. Narrative literature review

In the extraction step, REE-bearing material undergoes several pretreatment options, such as crushing, grinding, milling, incineration, combustion, and smelting, depending on the feedstock type. The extraction step is intended to increase the REE concentration in the ore to at least 70% [51]. Then the purification step removes contaminants in the pretreated feedstock, which is essential in preparing the materials for the separation phase [3–5]. The separation step may include other substeps, such as the separation of heavy REEs from light REEs, or separating specific REEs, depending on the desired final product. The narrative literature review herein identifies various separation techniques (Fig. 2), explores basic concepts, defines chronological advancements, and identifies technological challenges reported in prior published studies.

2.1. Chemical approaches

Chemical separation approaches for REE separation have shown great promise and have successfully recovered REEs up to 99% purity [2]. However, these methods generally consume high volumes of chemicals. Table 2 presents a summary of the advantages and disadvantages of the leading chemical separation approaches. Hydrometallurgical operations are an essential part of extractive metallurgy and are utilized in various metal refining facilities throughout the world [52]. They are known to be flexible and highly selective in the treatment of raw materials for metals. During hydrometallurgical treatment, the main processes include leaching, solvent extraction, ion exchange, and precipitation, which vary depending on the material being recovered [52]. In the separation phase, REE-bearing alloys (after initial purification to remove contaminants) are dissolved in powerful mineral acids comprising any or all of the four major stages, i.e., leaching, solvent extraction, ion exchange, and precipitation [53–55].

2.2. Physical and thermal approaches

Physical and thermal separation approaches are typically paired with two main mechanical recovery methods, including (i) pre-consumer recycling, which involves recycling performed on manufacturing scrap, and (ii) post-consumer recycling, which entails recycling discarded REE-bearing consumer products [51,64]. For metals, recycling usually has three major steps: collection, pre-processing (pretreatment), and processing. Discarded products first undergo cleaning in the collection step, followed by a pre-processing step where discarded items undergo physical dismantling [64,65]. The processing step uses pyrometallurgical techniques, such as smelting in furnaces, incineration, combustion, and pyrolysis, to reduce undesired products into particulates for downstream separation of REEs [51,54]. Globally, the pyrolysis and smelting route is used about 70% of the time [66]. Table 3 summarizes the advantages and disadvantages of physical and thermal REE separation approaches after size reduction has occurred either through grinding, shredding, or pyrometallurgical processes.

2.3. Electrical approaches

The direct use of electrical current is another REE separation approach that can achieve highly selective and ultrapure REE products downstream. Unlike hydrometallurgical processes, electricity-aided separation of individual REEs is usually performed, using a combined pyrometallurgical and hydrometallurgical pathway. For example, the electrochemical approach integrates electro and hydrometallurgical methods, where both metal separation and metal electrowinning can be achieved in one setup [75]. Electrodialysis is another example of an electrochemical process in which the solution of interest is separated from impurities [76]. In this process, feedwater passes through a channel between two ion-exchange membranes, where a cation exchange membrane is only permeable to cations, and an anion exchange membrane is only permeable to anions. The driving force for this movement is an electrical field created by two electrodes. The negative electrode attracts the cations, whereas the anions are attracted to the positive electrode. The flow passing through the ion exchange membranes has a high ion concentration, so the feed salinity decreases, and desalination occurs [76]. Table 4 presents an overview of the advantages

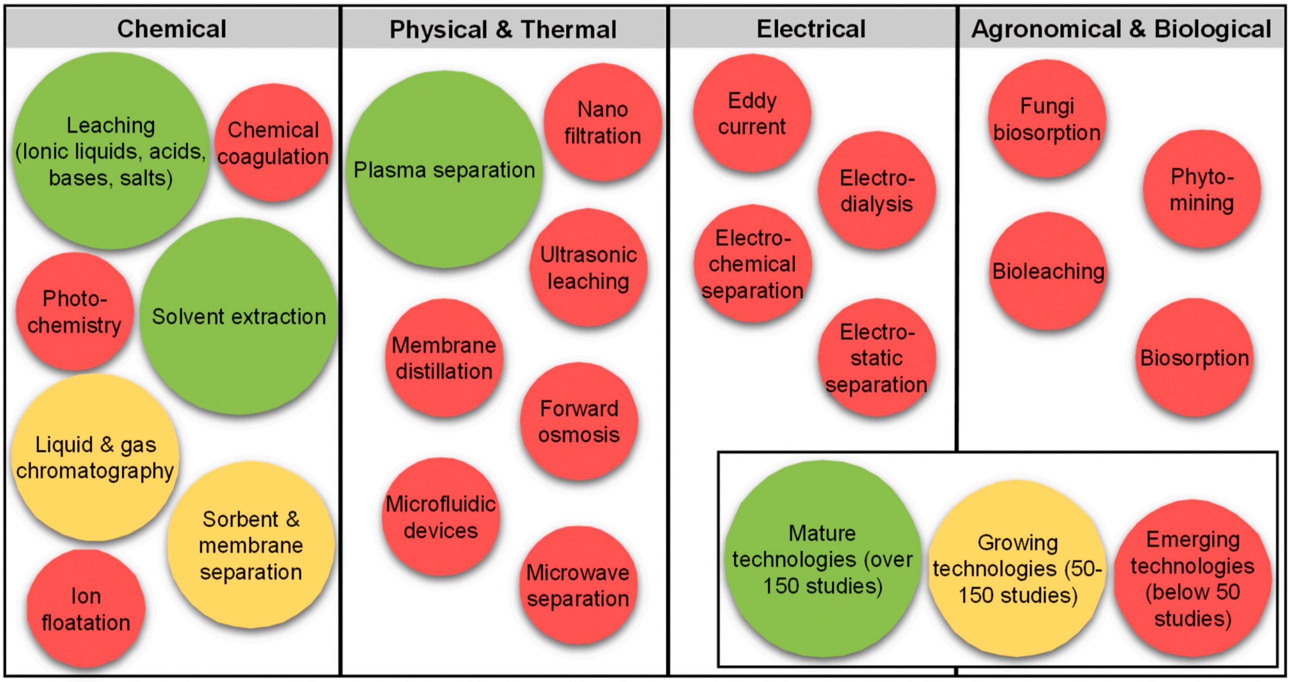

Fig. 2. Recent mature, growing, and emerging REE separation technologies (from Jan. 2015 to Jan. 2020).

Table 2
Chemical approaches for REE separation.

Method [Ref.]	Mechanism	Advantages	Disadvantages
Leaching [56]	Extracting metals into solution, using special reagents (acids, bases, or salts) followed by separation by filtration, precipitation through solvent extraction, and recovery by hydrometallurgy.	• High REE selectivity • Easy to perform • Low energy demand • Used commercially	• Requires a large amount of chemical reagent • Difficult to recover REEs at low concentration • Environmentally hazardous tailings
Photochemistry [57,58]	REE separation based on its light absorptive properties, using visible light, ultraviolet, or infrared radiation. Solvents and additives are used to control the charge transfer band, changing required light wavelengths.	• Low cost and emission (hazards) • High recovery efficiency and purity	• High dilute solutions use • Expensive to use light in high intensities
Sorbent and membrane [59]	Selectively recovering targeted REEs, using a membrane with different pore sizes and adsorbing capacities.	• Adjustable shape and size to selectivity separate targeted REEs • High recovery purity • Low chemical usage and waste generation • Low cost and energy consumption	• High membrane replacement cost • High chemical reagents used for cleaning
Liquid and gas chromatography [60]	Particles pass through a medium and are separated by the speed at which they pass.	• High separation resolution for REE ions • High separation efficiency	• High capital and operational costs
Chemical coagulation [61]	Bringing non-settling particles together into larger/heavier solids masses (floc), using chemical coagulants (e.g., metallic salt) added to water. A precipitant can be added to facilitate bonding of precipitates before coagulant is added. A coagulant oppositely charged to the particles is added, and charges are neutralized. Van der Waals forces then cause the particles to floc together.	• High removal capacity	• High operational cost • High chemicals use and disposal
Ion flotation [62,63]	Removal of hydrophobic ions in a solution by air bubbles, using surface-active agents (surfactants) acting as collecting agents. The surfactant has a charge opposite to the ions being recovered. Gas bubbles carry ions to the surface, where they are collected in the foam.	• Low energy requirements and high applicability • High concentration of desired elements in produced sludge	• Low efficiency in separating complex mixtures
Solvent extraction [17,39,62]	Generally, follows leaching and involves separating, using an aqueous phase (e.g., acid load with REEs) and an organic phase in contact with each other.	• Effective in removing metal ions in water • Produces high purity single rare earth solutions and compounds • Used commercially	• Only applied to non-dilute metal ion solutions • Inefficient, time-consuming, and labor-intensive • High solvent requirement

and disadvantages of REE separation, using direct electricity-aided methods to separate REEs after undergoing extraction and concentration, and preliminary removal of impurities for selective REE recovery.

2.4. Agronomical and biological approaches

Agromining processes involve growing hyperaccumulator plants as a crop, then harvesting the plant, drying, ashing, and processing it to

Table 3
Physical and thermal approaches for REE separation.

Method [Ref.]	Mechanism	Advantages	Disadvantages
Plasma [67,68]	Heating materials to very high temperatures to break bonds until atoms are ionized, leaving individual ions and electrons. Specific elements are then extracted from the bulk and either recombined or deposited on surfaces.	• No secondary waste streams • A single-step process • No chemical requirements • Environmentally friendly	• High cost for streams with low REE concentration • High energy requirement
Microfluidic device [69]	Selectively separating REEs, using a microfluidic chip in solvents.	• High separation efficiency for heavy REEs after 10 s in mixed REE oxide	• Inadequate phase separation times in complex REE extraction circuits • High energy requirement • Dilute solution need
Membrane distillation [62,63,70]	Separating desired elements, using a hydrophobic membrane and heat.	• No chemical consumption • High metals concentration • Low operating temperature	• High cost and membrane replacement
Ultrasonic leaching [71,72]	Extraction of REEs in solution, using an ultrasound processor, breaks apart particles and chemical bonds under microscale cavitation.	• High penetration • High separation efficiency due to cavitation effect	• High energy and maintenance requirements
Nanofiltration [62]	REE separation, using size-exclusion membranes or size discrimination membranes.	• High metal separation efficiency • Efficient passage of mono-charged ions	• High cost and membrane replacement • Low permeate flux
Microwave [73,74]	REE separation, using an aqueous solution of REE and a non-contact and on/off heating technique.	• Efficient heating to 1000 °C in a few seconds • High material selectivity • Dielectric characteristics reduce heating time • Superior absorption characteristics of the aqueous solution	• Insoluble large particle sizes after microwave roasting

recover targeted REEs [82,83]. The use of plants and fungi to extract and concentrate metals, also known as phytoextraction, has been studied by several investigators, and includes phytoremediation and phytomining methods [84–86]. Phytoremediation is the process of recovering metal contaminants (e.g., cadmium and lead) in the soil for safe disposal [87], while phytomining uses plants to recover REEs and valuable metals, such as gold, platinum, and thallium [84,86]. The hyperaccumulation process occurs naturally for many metals that can be induced in some

Table 4
Electrical approaches for REE separation.

Method [Ref.]	Mechanism	Advantages	Disadvantages
Electrochemical [75,77,78]	REEs extraction in solution or an electrochemical cell, using integrated electrowinning and hydrometallurgy.	• Low chemical reagents use and high base metals recovery • Less energy-intensive relative to pyrometallurgy	• Leaching solution requirements • Decreased efficiency over time
Electrostatic [51,66,79]	Selectively separating particles based on surface charges, using the electrical conductivity of charged particles.	• High separation efficiency for small particles (100 μm-10mm)	• High intermediate products • High impurity of nonconductive products
Electrodialysis [70,76,79,80]	REEs separation through the flow of ions in a semi-permeable membrane with the aid of an electric potential.	• Low pressure (~15 psi) • Eliminating impurities through an ion-exchange membrane	• High operation cost for salinity greater than 5,000 ppm
Eddy current separators [81]	Separating metals based on their conductivity and density, using a magnetic field.	• High separating efficiency for ferrous metals, non-ferrous metals, and non-metallic	• Challenging to achieve separation between REEs

plant species by adding chemicals to solubilize metals for plant uptake [82]. The metal uptake process through plants occurs in six steps: (1) solubilization of metal from the soil matrix [88], (2) acidification of the rhizosphere [89], (3) secretion of ligands by the rhizosphere of the microorganisms [82], (4) root absorption and transport to plant shoots, (5) distribution, and (6) sequestration of the metal ions [58,59]. Various oxidation states of heavy metals have different uptake, transportation, and detoxification characteristics in plants. Once the metals are translocated to shoot cells, they can be stored in cellular locations, such as trichomes (apoplast tissue), epidermis, mesophyll, and cell wall, where the metals will not damage vital cellular processes [90]. The final step for accumulating most metals is sequestration that usually occurs in the plant's vacuole, where the metal or metal-ligand is transported across the vacuolar membrane. Table 5 provides a summary of the advantages and disadvantages of REE separation, using agronomical methods.

In cases where pyrometallurgical or hydrometallurgical approaches are not cost-effective or environmentally friendly, unicellular microorganisms (e.g., fungi or bacteria) [94], or microbiological products provide suitable alternatives for biomining [38]. Biomining includes two main steps: bioleaching and bio-oxidation [95]. Bioleaching is commonly applied for REEs and precious metals separation [95]. Particularly, heterotrophic organisms consume organic carbon sources and produce organic acids for the metal leaching [38]. These organic acids promote the dissolution of solids and have chelating properties that improve REE solubility [38]. Additionally, bioleaching is performed by other microorganisms with the ability to excrete organic acids (e.g., citric or gluconic acid) or aid in the production of inorganic acids (e.g., cyanide or sulfuric acid) for dissolution or oxidation and reduction of metals [96–99]. Acidophilic sulfur-oxidizing bacteria and iron-oxidizing bacteria are the most widely used [97,100]. The main bioleaching process steps for REEs separation from e-wastes are acidolysis, redoxolysis, and complexolysis. In acidolysis, the microbial acids (e.g., malic, gluconic, or sulfuric) cover the surface of the e-waste [101]. During redoxolysis, microbial growth occurs through electron transfers from metals, leading to the dissolution of the metal [97,100,102]. Finally, cyanogenic bacteria and fungi separate the targeted metals during complexolysis. Table 6 provides a summary of the advantages and

Table 5
Agronomical approaches for REE separation.

Method [Ref.]	Mechanism	Advantages	Disadvantages
Phytomining with shoots and leaves [83,84,91]	Metals separation, using the hyperaccumulating properties of different plants leave/shoot.	• Environmentally friendly • Low operational cost	• Plants need a constant nutrient supply • High land requirement
Phytomining with root crop [83,84,91]	Metals separation, using the hyperaccumulating properties of various crops root.	• Environmentally friendly • Low operational cost	• Crops need nutrient supply High land requirements • Non-selective REE hyperaccumulation
Phytomining with mushroom [85,92,93]	Metals separation, using the hyperaccumulating properties of different mushroom species.	• Low cost • Environmentally friendly	• Mixed REEs hyperaccumulation vs. fractionated REE

Table 6
Biological approaches for REE separation.

Method [Ref.]	Mechanism	Advantages	Disadvantages
Fungi (unicellular) biosorption [101]	Accumulating metals with high concentrations, using fungi.	• Applicable in high pH environments • Low operational costs compared with conventional ore mining methods	• Long process time
Bioleaching, biometallurgy, or biomining [52,103,104]	Extracting valuable metals from low-grade ores, using microorganisms (bacteria or archaea)	• Environmentally friendly • Low temperature and energy requirements • Appropriate for treating low grade and waste metals • Low operational costs	• Difficulty in microorganism reproduction • Bacteria toxicity • Low reaction rates • Large land requirements
Biosorption [94,98,105]	Binding and concentrating specific ions and molecules from aqueous solutions, using various properties of biomass or biomolecules	• Low operational costs • High metal removal efficiency • Environmentally friendly • Efficient removal in dilute solutions	• High adsorbent needs • Limited recovery

disadvantages of REE separation, using biological-based methods.

3. Systematic literature review

The systematic review conducted herein covers the current state-of-the-science for REE separation to maintain up-to-date knowledge of prior breakthroughs and identify the existing techniques in various fields. Systematic review studies aim to reduce bias from the authors that usually reinforce partialities and their research interest. Also, systematic reviews aid in determining the critical parameters and key methodologies from prior published studies to guide future research and development by investigating the state-of-the-art in current and next-generation approaches and technologies. Fig. 3 presents the number of published studies during the last ten years, using only 'rare earth element' and 'separation' keywords in the Web of Science™ between January 2010 and August 2020. The results show a consistent growth in publication records, which is an indicator of increasing interest in the REE field. Due to the high number of publications, a detailed analysis was conducted, using keywords, topics, and abstract searches between January 2015 and January 2020. The keyword sets can be found in the Supplementary Material, Table S1. Fig. 4 presents the number of publications for each REE separation method in over 40 different fields of study during the last five years, as well as the research stage (i.e., early, emerging, and mature), using red and yellow (warm colors) for less frequent keywords and green (cold color) for more frequent keywords. The top three separation techniques are leaching, solvent extraction, and plasma with 1,121, 630, and 176 publication records, respectively. The dominant study fields are chemistry, engineering, and metallurgy with 697, 505, and 210 publication records, respectively.

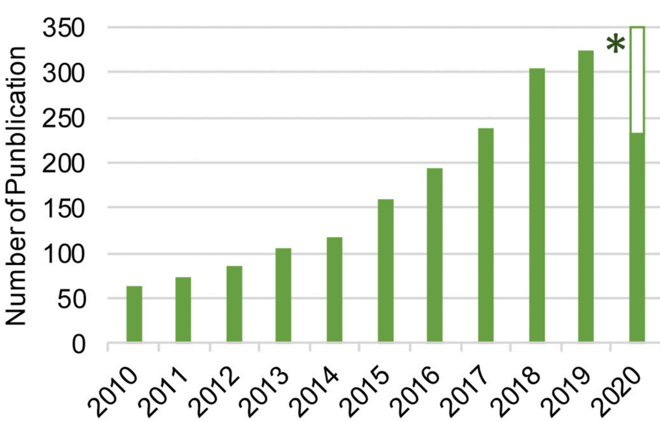

Fig. 3. Number of published articles on REEs separation between Jan. 2010 and Aug. 2020 (* estimated for Aug.–Dec. 2020).

Table 7 presents the top five countries and organizations with the highest publication records between Jan. 2010 and Aug. 2020. China, United States, and Japan are the top three countries with 669, 376, and 343 published articles. Chinese Academy of Sciences, Russian Academy of Sciences, and United States Department of Energy with 206, 160, and 100 publication records are the top three productive organizations during the last ten years.

4. Discussion

According to the United States Department of Commerce's Bureau of Industry and Security surveys in 2016 for determining the major challenges of the global REEs supply chain network, 81 (out of 160) REE producers (respondents) ranked 'Foreign Competition' as an organizational challenge, and 30 respondents claimed that it was their number one challenge [106]. The survey results show that other concerns to maintain competitiveness in the marketplace were frequently cited by all respondents, such as old, aging equipment, facilities, and infrastructure that will subsequently reduce the process efficiency and profitability. Due to these challenges and concerns, 66 respondents indicated that they imported most of their REE-related sources (e.g., ore, inorganic/organic purified compounds, and metals). Particularly, 46 of them imported from a single country (e.g., China or USA), and China was accounting for 28 (over 60%) respondents in 2016 [106]. Additionally, the results show that only 4% ($254 million) of the total budget has been applied for research and development in REE production

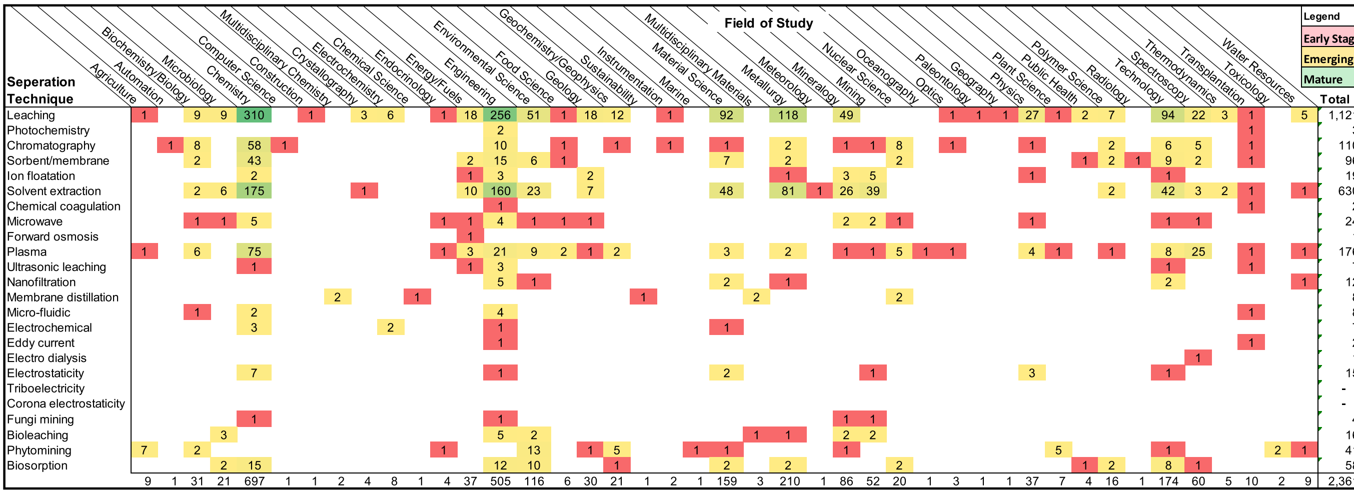

Fig. 4. Systematic review for REE separation techniques between Jan. 2015 and Jan. 2020.

Table 7
Top five countries and organization based on the publication records during the last 10 years.

Countries	Publications	Percentage	Organizations	Publications	Percentage
China	669	22%	Chinese Academy of Sciences	206	7%
USA	376	12%	Russian Academy of Sciences	160	5%
Japan	343	11%	United States, Department of Energy	100	3%
Russia	234	8%	Centre National De La Recherche Scientifique	87	3%
India	223	7%	Bhabha Atomic Research Center	61	2%

[106].

To overcome the 'Foreign Competition' challenge, Japan that was heavily dependent on China's REE export, swiftly turned to its urban mines (e.g., municipal landfills) and e-waste for addressing their REEs needs. According to recent estimates from the Japanese National Institute for Materials Science, their urban mines held approximately 300,000 tons of REEs [49,107]. This discovery encouraged Dowa Holdings, a former mining company from Kosaka, Japan, to build a recycling facility, which melts old electronics and extracts valuable rare metals from the molten stew [49]. So far, the recycling facilities successfully recover rare metals, for example, from liquid-crystal display units and antimony (Sb) from semiconductors [108]. REE recovery from landfills can also mitigate negative effects on the environment and replenish the resources required to meet current and future technological demands [3–5]. E-waste recycling is beginning to gain momentum in the United States. Recent studies show that approximately 25% of 1.8 million metric tons of domestic e-waste were recycled or reused in 2018 [27,109]. However, a cost-competitive, environmentally friendly, and scalable approach has not been achieved for REE recycling yet. Despite the many advances made so far, most REE technologies exist in the lab-/pilot-scale rather than on a commercial scale [28], which is why several countries export their e-waste to developed countries for recycling.

Currently, leaching and solvent extraction are two main highly applied and mature REE chemical separation techniques. Leaching is a simple process, requiring low energy and providing high selectivity of REEs; however, the main drawbacks are high chemical reagent needs, low efficiency for separating low REE concentrations, and the hazardous byproducts [56]. Solvent extraction is an effective separation technique that is commercially used and produces high purity single REE solution or mixed REE compounds; however, it does struggle from inefficiencies, being labor-intensive, and time-consuming [17,39,62]. Other chemical approaches are immature and growing technologies, such as photochemistry, sorbents/membranes, chromatography, chemical coagulation, and ion flotation. Photochemistry can be performed with high efficiency and purity at relatively low costs with little emissions; but, it is performed in dilute solutions (low concentrations) and can be expensive to use if high-intensity light is required [57,58]. Membranes have the advantage of low chemical use and waste generation, however, the cost of replacement membranes can be high, and cleaning requires hazardous chemicals [59]. Chromatography can be expensive to use for separating REEs, but it does provide high separation efficiency and resolution for REE ions [60]. Chemical coagulation can remove high amounts of REE from the solution, but the high operational costs and large amount of chemicals needed make it commercially ineffective on a large scale [61]. Ion flotation has low energy requirements and high recovery efficiency, however, this technique suffers when trying to separate complex mixtures [62,63].

Prior physical and thermal separation approaches show plasma separation is the only highly applied and mature technology, and other techniques (e.g., microfluidic, membrane distillation, ultrasonic leaching, microwave, and nanofiltration) are still immature and growing for REE separation. Plasma separation is an environmentally friendly, single-step process with no chemicals requirement and very low waste streams; however, it needs high energy requirements and is expensive for streams of low REE concentration [67,68]. Microfluidic devices have high mass transfer coefficients and short mass transfer distances, giving them high separation efficiency of REEs at low concentrations. These devices suffer from high energy demand and long phase separation times in complex solutions [69]. Membrane distillation can separate REEs without using chemicals under low temperatures and works for highly concentrated solutions; however, the cost of membrane replacement is its main drawback [62,63,70]. Ultrasonic leaching has shown overall higher separation efficiency in comparison to traditional leaching due to the effects of cavitation, but it still requires high energy and constant maintenance to carry out [71,72]. Nanofiltration allows mono-charged ions to pass through while rejecting multi-charged REE ions for efficient separation; however, it suffers from high-cost membranes and low permeate flux, requiring long process times [62]. Microwave separation is a newer technique and has many good qualities, such as quick, efficient heating, high selectivity, and superior absorption characteristics. There are large insoluble particles that are produced from microwave roasting, which can affect the efficiency [73,74].

Most electrical separation approaches (e.g., electrochemical, electrostatic electrodialysis, and eddy current) are still growing and require further investigation. Electrochemical separation requires a relatively small amount of chemical reagents for high metal recovery and low energy requirements. It is generally used in conjunction with leaching and not as a stand-alone process. Also, the efficiency can decrease over time due to the coating of the cathode [75,77,78]. Electrostatic separation can separate small REE particles with high efficiency, but produces a large amount of intermediate products and impure nonconductive products [51,66,79]. Electrodialysis uses mild operating conditions and can eliminate impurities by ion exchange; however, the operation cost significantly increases depending on the needed salinity amount [70,76,79,80]. While eddy current separators have high efficiency for various metals, but it still needs further improvements to address multiple challenges for the separation of different REEs [81].

The trend of e-waste exports could be further reduced with new technologies and approaches (e.g., agromining) [110]. Agromining is a single-step REE extraction, pre-concentration, and separation approach that can be designed to be feedstock agnostic. Besides, agromining can be useful in enabling small to medium size companies and municipal landfills in the United States and developing countries to participate in REE recycling at a fraction of the cost of ore mining [83,110]. Particularly, pyrometallurgy and hydrometallurgy are unsustainable approaches due to high energy requirements and environmental impacts [111–113]. However, when the current mature REE separation technologies are coupled with the agromining approach, they could minimize the overall environmental effects and enhance sustainability benefits across waste-to-REEs life cycles. The agromining processes employ perennial plant species with high hyperaccumulation characteristics, such as rapid regeneration above-ground after harvesting, and continuous or year-round production of metal-rich bio-ore [114]. After the REEs have been pre-concentrated in the plant or the body of multi-cellular fungi (e.g., mushrooms [92]), REE separation into individual elements can be achieved through several hydrometallurgical [115], or electrochemical processes [75,116]. Utilization of the hyperaccumulating characteristics of plants for phytomining makes it an environmentally friendly and low-cost process, but it requires a lot of lands and constant plant care.

Currently, biological separation approaches (e.g., biosorption and bioleaching) are immature and need further advancements. The ability of microorganisms to change the speciation of metal allows for environmentally friendly REE separation. For example, using fungi as the organism enables the cultivation of REEs in high pH environments at low operational costs, but the main downside is the low process rates [101]. The use of microorganisms for bioleaching results in low emissions, energy requirements, and operational costs. All of which are advantageous compared to current methods. However, microorganism production and the slow reaction rates, as well as large land requirements, keep bioleaching from being feasible [52,103,104]. Biosorption has many advantages, such as low operational cost, lack of hazardous waste, high metal removal efficiency, and high efficiency to remove particles in dilute solutions. However, biosorption suffers from limited REE recovery and has high adsorbent requirements [94,98,105].

Grosjean et al. (2019) conducted a study under hydroponic conditions and examined five Phytolacca species (i.e., americana, acinosa, clavigera, bogotensis, and icosandra) for REEs recycling [110]. They observed that all the plant species indicated a little or no oxidative stress after accumulating REEs, and heavy REEs were preferentially transferred to plant leaves. Furthermore, Dicranopteris dichotoma and Pronephrium simplex were also observed to have the highest REE hyperaccumulation rates among ferns with REE concentrations of 0.7% dry weight of light REEs and 1.2 g REE/kg dry weight. In comparison to other REE-accumulating species, they reported that Phytolacca americana (American pokeweed or dragonberries) was the fastest-growing plant, reaching to 3 m in height [110]. Also, Robinson et al. (1997) observed that other hyperaccumulators (e.g., berkheya coddii, alyssum bertolonii, haumaniastrum robertii, haumaniastrum katangense, thlaspi caerulescens, thlaspi calaminare, alyssum lesbiacum, artemesia californica, and brassica juncea) have high metal absorption properties [88]. They concluded that Berkheya coddii had several advantages over other candidates due to their ease of growth from seed, tolerance to cool climate conditions, prolific reproduction of seeds, resistance to insect and soil pathogens, and their accumulation of metals.

Similarly, Borovička et al. (2011) investigated the occurrence and distribution of 16 REEs in edible saprobic mushrooms, such as Macrolepiota procera [117]. They observed that the mean concentration of 14 REEs (i.e., La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, and Lu) was 0.50 and 0.75 mg per kg of dry biomass in the caps and the whole fruiting bodies of the fungi, respectively. Additionally, they reported that Ce was the most abundant REE in edible caps of the mushroom with a mean concentration between 0.18 ± 0.29 mg per kg of dry biomass. This phenomenon was also observed by Haselwandter et al. (2011) in other mushroom species (i.e., Suillus granulatus, Suillus luteus, and Pleurotus ostreatus), which also showed the levels of REE concentration in their fruit bodies [118]. Several studies and investigators, such as van der Ent et al. (2015), proposed agromining methods (e.g., phytomining) for REEs recycling to provide local communities with an alternative type of agriculture on degraded lands [83]. In other words, farming not for crops, but metals, such as nickel. To improve the economic returns of degraded agricultural lands - ultramafic soils are deficient in calcium, potassium, phosphorus, excess magnesium, and nickel - a co-cropping approach could be used. In Greece, olive plantations were intercropped with Alyssum, and in Malaysia, palm oil estates were intercropped with Phyllanthus to improve the economic prospects of local farmers [83]. However, two decades after its inception and numerous successful experiments, commercial phytomining has not yet become a reality [114]. The slow or non-existent adoption of large scale agromining for demonstration purposes to identify operational risks and provide real-life evidence for profitability could be overcome when municipal landfills are used as mining sites [83]. Funari et al. (2016) reported that municipal solid waste incineration breaks REE-bearing solid waste into ashes for further treatments [119]. Rather than incineration, an energy-efficient sono-bioleaching process could be used to achieve size reduction and dissolution of REE-bearing e-waste for agromining to provide an environmentally friendly and profitable alternative [72]. In addition to urban mines, multiple resources could diversify the REEs recovery to ensure unhindered REEs supply and meet present and future needs, such as wastewater [36], coal fly ash [120], phosphate tailings [121], geothermal brines [122], seawater [108], meteorites [123], and extraterrestrial planetary bodies [114,124].

5. Conclusions and future directions

Over the past two decades, REEs consumption in various sectors (e.g., aerospace, healthcare, transportation, and communications) have gained significant attention due to growing global shortage and supply risks. These concerns have accelerated research and development in finding new or integrated approaches, as well as identifying other sources (e.g., e-waste) to support a competitive, sustainable REE supply. However, there is a shortage of literature, including a detailed assessment of each individual and integrated processes for REE production or recycling from various resources, such as ore mines and municipal landfills. This study provides a comprehensive overview of existing REE separation techniques, process challenges, and potential science and engineering opportunities through both narrative and systematic literature reviews. The narrative review examines the current studies and identifies the possible pathways and technologies to bridge existing gaps and future perspectives across REE production and recovery practices. It is evident that the existing REE production techniques are extraction/pre-concentration, purification, separation, and refining, and the developed techniques can be classified into four primary approaches, which are chemical, physical/thermal, electrical, and biological/agronomical. From the systematic review, it is apparent that REE production and recovery practices have been a rapidly growing field over the last ten years. From both narrative and systematic reviews, it is clear that chemical and electrical/thermal approaches (e.g., leaching, solvent extraction, and plasma separation) are the most explored and mature separation approaches for REE recovery from end-of-life products.

As of yet, a cost-competitive and environmentally friendly approach for REE separation has not been achieved; therefore, the opportunities remain for exploring either new or mixed techniques to enhance the separation efficiency and sustainability benefits. At present, the need for the process integration across fields of knowledge is essential to reveal the gaps between the lab-scale studies and industry practices. This study directs future research towards developing sustainable REE production from multiple feedstocks by integrating agromining approaches (e.g., phytomining) with the predominantly explored techniques (e.g., leaching) to address the stated sustainability and commercialization challenges. Particularly, the improved REEs concentration in hyperaccumulators would shorten the overall REE production cycle from mining to REE bio-ore production, and improve annual production throughput. A fully integrated and scalable REE agromining system can be designed to be feedstock agnostic, yet able to fractionate high purity REE concentrates, and enable companies and municipal landfills to simultaneously mitigate the environmental impacts of urban wastes, while producing REEs to meet the future demands. Further investigation to advance REE separation techniques are as follows:

- ➢ Exploration of agromining hyperaccumulators (e.g., Phytolacca americana or Phytolacca acinosa) that can fractionate light REEs from heavy REEs in their plant
- ➢ Exploration of agrophysics techniques (e.g., manipulating magnetic fields, sonoporation, and light intensities) that can enhance REE concentration in growing hyperaccumulator plants

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.rser.2021.110917.

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